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(54) Benævnelse: **TERMOPLASTISK SAMMENSÆTNING**

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The present invention relates to a thermoplastic composition, such as is used in particular in extrusion and compounding, as well as to a method for producing foils, films, shaped articles, foam materials, coatings, paints and dispersions using the thermoplastic composition according to the invention, as well as foils, films, shaped articles, foam materials, coatings, paints and dispersions produced using the method according to the invention.

Thermoplastic compositions, particularly those involving biogenic polymers, are known in the state of the art and are used in particular in extrusion and compounding. State of the art methods for producing polymer blends usually work according to the basic scheme that a polymeric carrier material as the main component of a formulation and one or more other polymers are extruded together. Here, very often fillers are usually added in the upper area of the feed zone of the extruder in order to vary the specific properties of the product and at the same time reduce the formulation costs. The purpose of further additives is to improve the compatibility between the polymeric starting materials and the fillers. After all, plasticisers are indispensable in many formulations, both for process control and for the product result.

A stable and robust process control is of crucial importance for an optimal result, whereby the percentage proportion of water in the compound is of crucial importance.

Water can be used as a formulation component in compounding and extrusion processes and especially in the function as a plasticiser. However, the use of water has in particular the disadvantages described below.

Extrusion processes usually take place well above the boiling point of water, such that, if the degassing is incomplete, the water-containing compound will foam uncontrollably. At the same time, the extrusion processes take place under high pressure. The pressure drops abruptly in the area of the outlet nozzle of the extruder. This means that there is a sudden expansion of the extrudate water, and thus an increased foaming of the extrudate at the outlet nozzle begins, with hindering effects on the extrusion result and the further process control. Efficient degassing, for example by vacuum extraction, can potentially counteract this effect, but is associated with considerable technical effort.

Furthermore, under typical extrusion conditions at zone temperatures of >>130°C, which are usually required for such compositions, water can already act as an electron donor (Lewis base), since it has anhydrous valences which are suitable for the formation of covalent bonds and thus trigger disadvantageous nucleophilic reactions. The hydrolytic degradation of polyesters is a well-known disadvantageous effect in the state of the art, wherein, in addition to the actual degradation reaction, undesirable postpolymerisations and crosslinking reactions can take place. In the case of typical biogenic polymers such as polysaccharides, hemicelluloses or polyhydroxyalkanoates, but also petroleum-based biodegradable polymers such as e.g. polyvinyl derivatives, this leads to cleavage reactions, the formation of small molecular fragments, and transesterifications, so that the extrusion process as a whole can become uncontrollable. In addition, water undesirably forms azeotropic blends with many small molecules of different polarity, in which the vapour and liquid phases are the same. Their composition has uncontrollable effects on the compounding and extrusion processes. Of course, water can be trapped by using crosslinking anhydrides such as maleic acid, succinic acid, phthalic acid, caprolactam. However, this inevitably leads to potentially disadvantageously altered material properties because the anhydrides are converted into chain extenders or crosslinking agents.

With many biodegradable and/or compostable plastics, however, a significantly high disadvantageous water content is often unavoidable. With these plastics, for example, starch is often added in native form as a structural element, which replaces conventional non-biogenic structure formers. About 12-20 % of water is a structural component of native, macromolecular starch. Even with modified starch forms, intrinsic water values > 5 % are usually still achieved. The incorporation of starches into the compound poses a problem due to the relatively high intrinsic water content of the starches in the compounding process, unless the evaporation can be prevented by vacuum suction. However, the often incomplete degree of digestion of the specific starches can also lead to uncontrollable results.

Furthermore, starch under the influence of heat, such as occurs during extrusion, can physically bind, swell and gelatinise many times its own weight in water. When heated with water, the starch swells by storing water at approx. 47 - 57 °C, and at approx. 55 - 87 °C so-called starch paste is formed by starch digestion, which has a different viscosity depending on the type of starch. The starch that has already gelatinised, can support the gas retention in the extrudate and lead to solid pores with corresponding undesirable consequences for the further process control.

In order to effectively process the starch well during extrusion and compounding in the case of starch-containing biodegradable and/or compostable plastics, a plasticiser must be used. Numerous plasticisers are known in the state of the art that are used for compounding and extrusion processes. Furthermore, in the state of the art WO 99/40797A1, US 2013/136879 A1, US 2003/166748 A1 and US 2011/311743 A1 are known that relate to the technological background of the present invention.

It is an objective of the present invention to at least partially overcome the disadvantages known from the state of the art. The above objective is achieved by a thermoplastic composition according to the invention according to claim 1. In particular, fillers and/or plasticisers are used according to the invention not only for product variation and cost control, but rather as quenchers for trapping or inerting water during the compounding and extrusion process. Preferred embodiments of the thermoplastic composition are the subject-matter of the dependent claims.

The thermoplastic composition according to the invention for use in extrusion and compounding comprises:

(a) a continuous phase that has at least one first polymer which is selected from a group containing a polyester, a polyesteramide, a polyester etheramide, an aliphatic or partially aromatic polyester urethane or blends thereof, in particular such compounds or blends which have hydrophobic surface properties,

(b) at least one dispersed phase, which comprises

(i) as a second polymer, at least one biogenic crosslinking agent having a water content of 0.1 to 30 wt.%, in particular of 5 to 25 wt.% with respect to the total weight thereof, and wherein the first polymer of the continuous phase is not miscible with the second polymer of the dispersed phase, and

(ii) at least an inorganic filler and

(iii) at least one plasticiser.

The thermoplastic composition according to the invention is characterised in that the plasticiser contains diglycerol or blends with diglycerol and the inorganic filler contains phosphates of calcium and/or magnesium and has a proportion of 0.1 to 40 wt.%, based on the total weight of the composition and wherein the composition is biodegradable and/or compostable.

For the purposes of the present invention, “hydrophobic” is understood to mean the tendency not to bind water or not to temporarily physically adsorb and/or not to adsorb any further water molecules in hydrated form. In particular, the surfaces of the first polymer can be modified by hydrophobic coatings, primers or seals.

It has been found surprisingly that the use of such modified enantiomeric ratios of diglycerol in a thermoplastic composition can at least partially overcome the described state of the art problems in compounding and extrusion processes and in particular suppress the described uncontrollable nucleophilic reaction mechanisms caused by gaseous water in the compound. Furthermore, the azeotropic effect is suppressed after blending a water-containing compound with adsorbed glycerol, so that glycerol can be used according to the invention in combination with such modified enantiomeric ratios of diglycerol. It has been found surprisingly, for example, that with such a modified composition, glycerol itself can be added in a proportion of up to 60% as a plasticiser component in the thermoplastic composition without significant evaporation of the extrudate water with the described disadvantageous effects taking place, and in previous applications this is up to extruder zone temperatures of approx. 190°C. The use of such enantiomeric ratios of diglycerol also enables so-called polyglyceryols to be added to the compound, which are not defined compounds, but rather oligomers produced on the basis of the glycerol monomer with statistical chain length distributions. Here, polyglycerols from the trimer such as, for example, the glycerol trimer, tetramer, pentamer, hexamer, heptamer, octamer, entamer, decamer or multiples thereof can be used according to the invention. These are difficult to incorporate into the compounding process without the use of the enantiomeric ratio according to the invention of diglycerol due to their high viscosity. This is because they are liquids with very high viscosities, which make precise dosing almost impossible and which prevent solid mixing in the compound along the screw path of the extruder, even if the viscosity drops due to temperature.

According to a further preferred embodiment of the thermoplastic composition according to the invention, the plasticiser has a blend of

(a) diglycerol and triglycerol, or

(b) diglycerol and glycerol, or

(c) diglycerol, triglycerol and glycerol,

wherein the blend according to (a), (b) and (c) in this composition comprises at least between 25 and 75 wt.% of diglycerol with respect to the sum of

(a) the diglycerol and the triglycerol, (b) the diglycerol and the glycerol or (c) the diglycerol, glycerol and triglycerol.

According to a further preferred embodiment of the thermoplastic composition according to the invention, the inorganic filler has hydrophilic properties.

For the purposes of the present invention, "hydrophilic" is to be understood as the tendency to bind water or to physically attach it temporarily and/or to adsorb further water molecules in hydrated form.

The hydrophilic inorganic fillers according to the invention therefore advantageously act as quenchers of the water of the thermoplastic composition and thus counteract the uncontrollable nucleophilic reaction mechanisms caused by gaseous water in compounding and extrusion processes, in particular when extruding in the region of the outlet nozzle of the extruder.

In the case of the inorganic fillers according to the invention, the surfaces in particular, but also internal components, can have hydrophilic properties. In particular, the surfaces of the inorganic fillers can be modified by hydrophilic coatings, primers or seals.

According to the invention, the inorganic filler comprises phosphates of calcium and/or magnesium.

The use of these fillers has a further advantage, which is that they can be used to produce elasticity values comparable to those of talc. This fact is most interesting because the carcinogenic potential of talc is classified as comparable with asbestos.

As a result, compared to a composition without such inorganic fillers, a higher elongation at break and tear propagation of the material is achieved, in particular when using sulphate salts of an alkaline earth metal. Furthermore, it has surprisingly been found that, in particular, sulphate salts of an alkaline earth metal, alone and particularly in combination with diglycerols in enantiomeric ratios according to the invention, are suitable for improving the compounding process and can also reduce or prevent the problems of blistering and foaming at the outlet nozzle of the extruder during extrusion.

According to a further preferred embodiment of the invention, the salts of the alkaline earth metals are calcium sulphates, preferably gypsum, such as, for example, natural gypsum, natural anhydrite, flue gas gypsum or blends thereof.

Gypsum in particular in combination with diglycerols in the enantiomeric ratios according to the invention is most advantageous for improving the compounding and extrusion processes.

According to a further preferred embodiment of the thermoplastic composition according to the invention, these are salts of the alkaline earth metals phosphate, preferably apatite, phosphorite and hydroxylapatite or salts and condensates of orthophosphoric acid, such as salts of pyrophosphates, polyphosphates and here in particular calcium polyphosphate, magnesium phosphate, sodium calcium polyphosphate, sodium magnesium polyphosphate and cyclophosphate of the alkaline earth metals magnesium and calcium.

A further preferred embodiment of the thermoplastic composition according to the invention, wherein the first polymer of the continuous phase

(a) has biogenic or partially biogenic components and/or is selected from a group of polyhydroxy acids, polyhydroxy alkanoates, polylactides and lignins and their derivatives and/or copolymers and in particular is selected from a group consisting of polybutylene adipate terephthalate (PBAT), polybutylene sebacinate terephthalate (PBST), polybutylene succinate (PBS), polybutylene succinate adipate (PBSA), and/or

(b) has synthetic components and is selected from a group of aliphatic and aromatic polyesters as well as aliphatic-aromatic copolyesters, for example polybutylene adipate terephthalate (PBAT).

According to a most preferred embodiment of the thermoplastic composition according to the invention, the biogenic or partially biogenic component or the synthetic component is polybutylene adipate terephthalate (PBAT) or has polybutylene adipate terephthalate (PBAT).

According to a further most preferred embodiment of the thermoplastic composition according to the invention, the biogenic component (a) is polybutylene sebacinate-co-terephthalate (PBST) or has polybutylene sebacinate-co-terephthalate (PBST).

According to a further most preferred embodiment of the thermoplastic composition according to the invention, the biogenic component (a) is polybutylene succinate (PBS) or has polybutylene succinate (PBS).

According to a further most preferred embodiment of the thermoplastic composition according to the invention, the biogenic component (a) is polybutylene succinate adipate (PBSA) or has polybutylene succinate adipate (PBSA).

According to a further preferred embodiment of the thermoplastic composition according to the invention, the composition comprises, as a further dispersed phase, at least one amorphous or semicrystalline and in particular biogenic polymer.

For the purposes of the present invention, "semicrystalline" is understood to mean a solid which has both crystalline and amorphous regions (domains).

According to a further preferred embodiment of the thermoplastic composition according to the invention, the amorphous or semicrystalline and in particular biogenic polymer is a polymer or copolymer of polylactic acid (PLA) or a polymer or copolymer of polyhydroxy alkanoates (PHA) and here in particular of polyhydroxybutyrate (PHB), polyhydroxyvalerate (PHV), polyhydroxybutyrate valerate (PHBV), polyhydroxy-butyrate hexanoate (PHBH), polyhydroxyhexanoate (PHH), polyhydroxyoctanoate (PHO) or a blend of at least two of the aforementioned.

According to a further preferred embodiment of the thermoplastic composition according to the invention, the biogenic crosslinking agent of the dispersed phase comprises polysaccharidic structural elements which originate from a group which includes starches, each of which in particular contain a specific composition of amylose and amylopectin such as, for example, potatoes, corn, buckwheat, wheat, rye, oats, barley, rice, peas and tapioca.

"Starch" here means all types of starch, i.e. flour, natural starch, chemically and/or physically modified starch, hydrolysed starch, destructured starch, gelatinised starch, plasticised starch, thermoplastic starch and blends thereof.

Starches that are completely destructured or are capable of being easily largely destructured have proven to be most advantageous.

Advantageously, the biogenic crosslinking agent of the dispersed phase of the present invention comprises more than one polysaccharidic structural element. Mixtures containing starch and at least one additional polysaccharidic structural element as biogenic crosslinking agents of the dispersed phase are most preferred.

In a preferred embodiment of the thermoplastic composition according to the invention, in addition to starch, the biogenic crosslinking agent of the dispersed phase has at least one additional polysaccharidic structural element, in particular selected from the group consisting of cellulose, hemicellulose and/or derivatives thereof, in particular acetate derivatives, ester derivatives and ether derivatives.

According to a further preferred embodiment of the thermoplastic composition according to the invention, the ratio of the polysaccharidic structural elements of the dispersed phase to the at least one first polymer of the continuous phase is 10 to 90 wt.% to 90 to 10 wt.%, preferably in a ratio of 30 to 70 wt.% up to 70 to 30 wt.% and more preferably in a ratio of 60 to 40 wt.% up to 40 to 60 wt.%, based on the sum of the polysaccharidic structural elements of the dispersed phase to the at least one first continuous phase polymer.

According to a further preferred embodiment of the thermoplastic composition according to the invention, the composition additionally has further additives selected from the group consisting of dispersants, lubricants, water-protective agents, crosslinking agents, phase mediators, mould removal aids, blend improvers, UV stabilisers and catalysts, as well as antioxidants.

Chain extenders such as mono-, di- and polyepoxides, polyepoxide-acrylates and their copolymers with styrene, aliphatic, aromatic or aliphatic-aromatic carbodiimide oligomers and polymers, isocyanates, isocyanurates and combinations thereof, anhydrides and polyanhydrides, which improve the compatibility between the polysaccharide structural element or the polysaccharidic structural elements and the other polymers of the composition, can also be added.

Furthermore, it is within the meaning of the present invention to use the various compositions according to the invention as master batches in state of the art methods.

A method for producing foils, films, shaped articles, foam materials, coatings, paints and dispersions using the thermoplastic composition according to the invention, is also subject-matter of the present invention.

The foils, films, shaped articles, foam materials, coatings, paints and dispersions produced by means of the method according to the invention are likewise subject-matter of the present invention.

Shaped articles within the meaning of the invention, such as plates, sheets, profiles and hollow articles, in particular bottles and containers, are known in the state of the art.

According to a preferred embodiment, the foils, films and shaped articles according to the invention have a wall thickness of between 5 μm and 1000 μm or a wall thickness of between 10 μm and 2000 μm.

The present invention also includes the use of the thermoplastic composition according to the invention, in particular in extrusion and compounding, for the production of foils, films, shaped articles, foam materials, coatings, paints and dispersions. The use of the thermoplastic composition in injection moulding, spraying, calendering, rotational moulding, dipping, painting, coating and pouring can also be within the meaning of the invention.

The invention is explained below on the basis of preferred example embodiments, wherein it should be pointed out that these examples also include modifications or additions which are immediately apparent to the person skilled in the art. Furthermore, these example embodiments do not represent a limitation of the invention in the sense that modifications and additions are within the scope of the present invention.

In the figures:

Fig. 1 shows the influence of the addition of calcium sulphate compounds, namely gypsum anhydride ($Ca[SO_4]$) (B), gypsum hemihydrate ($Ca[SO_4] \times {}^1/_2\ H_2O$) (C) and gypsum dihydrate ($Ca[SO_4] \times 2\ H_2O$) (D) compared to the addition of chalk ($Ca[CO_3]$, standard) (A) to the elongation at break of the composition according to the invention.

Fig. 2 shows the influence of the addition of calcium sulphate compounds, namely gypsum anhydride ($Ca[SO_4]$) (B), gypsum hemihydrate ($Ca[SO_4] \times {}^1/_2\ H_2O$) (C) and gypsum dihydrate ($Ca[SO_4] \times 2\ H_2O$) (D) compared to the addition of chalk ($Ca\ [CO_3]$, standard) (A) on the tear propagation of the composition according to the invention.

Fig. 3 shows the influence of the addition of calcium sulphate compounds, namely gypsum anhydride ($Ca[SO_4]$) (B), gypsum hemihydrate ($Ca[SO_4] \times 16\ H_2O$) (C) and gypsum dihydrate ($Ca[SO_4] \times 2\ H_2O$) (D) compared to the addition of chalk ($Ca[CO_3]$, standard) (A) on the breaking stress of the composition according to the invention.

Fig. 4 shows the influence of the addition of tertiary calcium phosphate ($Ca_3[PO_4]_2$) (B) compared to the addition of talc ($Mg_3[Si_4O_{10}(OH)_2]$, standard) (A) on the elongation at break (longitudinal and transverse) of the composition according to the invention.

Fig. 5 shows the influence of the addition of tertiary calcium phosphate ($Ca_3[PO_4]_2$) (B) compared to the addition of talc ($Mg_3[Si_4O_{10}(OH)_2]$, standard) (A) on the tensile strength (longitudinal and transverse) of the composition according to the invention.

Fig. 6 shows the influence of the addition of tertiary calcium phosphate ($Ca_3[PO_4]_2$) (B) in comparison to the addition of talc ($Mg_3[Si_4O_{10}(OH)_2]$, standard) (A) on the tear propagation (longitudinal and transverse) of the composition according to the invention.

Fig. 7 shows the influence of the addition of various proportions of talc ($Mg_3[Si_4O_{10}(OH)_2]$), namely 6% (A), 12% (B) and 18% (C) on the degradation of the composition according to the invention.

Fig. 8 shows the influence of the addition of various proportions of tertiary calcium phosphate ($Ca_3[PO_4]_2$) compared to the addition of 12% talc ($Mg_3[Si_4O_{10}(OH)_2]$, standard) (B) on the degradation the composition according to the invention.

Fig. 9 shows the influence of the addition of 6% secondary magnesium phosphate ($MgHPO_4 \times 3H_2O$) to 6% tertiary calcium phosphate ($Ca_3[PO_4]_2$) or 12% secondary magnesium phosphate ($MgHPO_4 \times 3H_2O$) in comparison to the addition of 12% talc ($Mg_3[Si_4O_{10}(OH)_2]$, standard) (B) on the degradation of the composition according to the invention.

Exemplary embodiments:

Formulations and process parameters with diglycerol

Variant 1

PBAT	Extruder	TSAEMP 26-40
Polylactide	D/L	26mm/40 D
Starch	Screw speed	200 [rpm]
Diglycerol	Pressure in front of the nozzle	14-15 [bar]
Chalk		
Talc		
Additives		

wherein the following can be used: PBAT in proportions between 30% and 70%, polylactide (dispersed phase) in proportions between 1% and 25%, starch in proportions between 1% and 25%, chalk (filler) in proportions between 1% and 25%, talc (filler) in proportions between 1% and 25% and additives in proportions between 0.1% and 5%, diglycerol between 1% and 6% based on the total weight of the formulation.

Zone	1	2	3	4	5	6	7	8 nozzle
Target	140	150	155	160	170	175	170	160
Actual	140	150	156	163	170	177	171	165

Variant 2

PBAT
Polylactide
Starch
Diglycerol
Glycerol
Talc
Additives

wherein the following can be used: PBAT in proportions between 30% and 70%, polylactide (dispersed phase) in proportions between 1% and 25%, starch in proportions between 1% and 25%, talc (filler) in proportions between 1% and 25% and additives in proportions between 0.1% and 5%, diglycerol and glycerol between 1% and 6% based on the total weight of the formulation.

Zone	1	2	3	4	5	6	7	8 nozzle
Target	140	150	155	160	170	175	170	160
Actual	140	150	155	165	170	175	171	162

Variant 3

PBST	Extruder	Leistritz ZSE 40
Polylactide	D/L	26mm/40 D
Diglycerol	Screw speed	180 [rpm]
Glycerol		
Chalk	Pressure in front of nozzle	11-12 [bar]
Talc		
Additives		

wherein PBST can be used in proportions between 30% and 70%, polylactide (dispersed phase) in proportions between 1% and 25%, chalk in proportions between 1% and 25%, talc (filler) in proportions between 1% and 25% and additives in proportions between 0.1% and 5%, diglycerol and glycerol between 1% and 6% based on the total weight of the formulation.

Zone	1	2	3	4	5	6	7	8	Flange 1	Flange 2	Nozzle
Target	135	135	140	155	155	155	155	160	160	160	165
Actual	135	135	135	155	155	155	155	155	160	160	165

In the case of a standard formulation, not according to the present invention, consisting of a matrix polymer and a further preferably biogenic polymer, the classic inorganic fillers are replaced by calcium sulphates, preferably gypsum. The present example not according to the present invention concerns the calcium sulphate compounds gypsum anhydride ($Ca[SO_4]$), gypsum hemihydrate ($Ca[SO_4] \times ½\ {}^{1}/_{2}H_2O$) and gypsum dihydrate ($Ca[SO_4]$ x 2 H_2O), wherein the following can be used: PBAT (continuous phase) in proportions between 50% and 70%, polylactide (dispersed phase) in proportions between 1% and 25%, calcium sulphate (filler) in proportions between 1% and 35%, talc (filler) in proportions between 1% and 25% and additives in proportions between 0.1% and 5%.

A	B	C	D
PBAT	PBAT	PBAT	PBAT
PLA	PLA	PLA	PLA
Ca[CO_3]	Ca[SO_4]	Ca[SO_4] x $^1/_2$ H_2O	Ca[SO_4] x 2 H_2O
(Chalk)	(Gypsum anhydride)	(Gypsum hemihydrate)	(Gypsum dihydrate)
Talc	Talc	Talc	Talc
Additive	Additive	Additive	Additive

It has surprisingly been found that the sulphates of calcium have a positive effect on the values of elongation at break and tear propagation. When using calcium sulphate, both elongation at break and tear propagation can be transversely improved by more than 10% (Fig. 1 and Fig. 2).

Not all parameters can be improved at the same time with the formulation according to the invention. When using sulphates in the compound, the breaking stress complementary to the elongation at break is reduced. However, the deviations of around 10% are relatively small (Fig. 3) and are therefore largely acceptable.

The positive effects of the composition according to the invention on the elongation parameters are very attractive for classic practical fields of application such as carrier bags, pouches or foils.

Variant 4

A compound consisting of a matrix polymer and a dispersed polymer phase is loaded according to the state of the art formulation (standard) with inorganic fillers such as chalk and talc for reasons of cost efficiency, but also to achieve specified strength values. As an example of the standard mentioned here, a proportion of 6%, 12% or 18% inorganic filler based on the total weight of the formulation has been used in the present case.

When replacing chalk or talc with tertiary calcium phosphate, it was surprisingly found that the degradation rate of the compound increased several times after just two weeks compared to the above-mentioned standard. For example, a foil according to the invention with 6% tertiary calcium phosphate reduced 1.4 times faster compared to a corresponding foil with talc (standard) instead of tertiary calcium phosphate, a foil according to the invention with 12% tertiary calcium phosphate reduced around 3.2 times faster compared to the standard (12% talc) and a foil according to the invention with 18% tertiary calcium phosphate reduced around 5 times faster compared to the standard (18% talc) (see Fig. 7, Fig. 8). It is noteworthy that the degradation rate of the foil according to the invention tends to decrease as the inorganic filler talc increases, but with the increase of the inorganic filler tertiary calcium phosphate increases, which enables a fine adjustment of the degradation rate by appropriate use of these fillers.

The degradation rate increases even more when the tertiary calcium phosphate is replaced or partially replaced by a secondary magnesium phosphate. In an example as described herein, the degradation rate multiplied 7-fold from 5% to 35% even if the proportion of tertiary calcium phosphate was halved from 12% to 6% and 6% secondary magnesium phosphate was used compared to a corresponding batch with talc (12%, standard; see Fig. 9).

However, if the proportion of secondary magnesium phosphate is increased to 12%, the degradation rate is already multiplied by almost 18. For the same reason, no further increase in the proportion of filler containing magnesium was tested in the aforementioned experiments. The degradation rate is then so high that the determination of material-specific properties with regard to their strength values hardly makes sense (not shown).

Apart from this last aspect, it is unexpected and advantageous that the material-specific properties of the tensile strength and elongation at break of the formulations according to the invention with tertiary phosphate-containing inorganic fillers and secondary magnesium-containing phosphates up to approx. 6% proportion differ only slightly compared to the talc-containing formulation standard (see Fig. 4, Fig. 5).

This also applies to the tear propagation (longitudinal), but not to the tear propagation (transverse) of the formulations according to the invention in comparison to the standard formulation (see Fig. 6). It is completely surprising that it has now also been found that the incorporation of phosphates into the formulation can dramatically improve the tear propagation in the transverse direction. This naturally decreases with increasing filler content, but is almost 300% higher than the talc-containing standard at 6% proportion, and still exceeds the talc standard by an enormous 120% even with 18% filler.

This is most noteworthy because, for example, in a film application after tearing, the tear propagation in the transverse direction is the decisive stability criterion.

The present formulations according to the invention thus allow advantageous control of the degradation behaviour of biodegradable compounds by varying the proportion of the formulation of inorganic phosphates while maintaining or even slightly improving the elasticity-defining parameters.

Patentkrav

1. Termoplastisk sammensætning, især til anvendelse i ekstrusion og kompoundering, omfattende:

(a) en kontinuerlig fase, som omfatter mindst en første polymer, som er valgt fra en gruppe, som omfatter en polyester, en polyesteramid, en polyesteretheramid, en alifatisk eller delvist aromatisk polyesterurethan eller blandinger deraf, især forbindelser eller blandinger, som har hydrofobe overfladeegenskaber,

(b) mindst en dispergeret fase, der

(i) som anden polymer omfatter mindst et biogent tværbindingsmiddel, som har et vandindhold fra 0,1 til 30 vægtprocent, især fra 5 til 25 vægtprocent i forhold til den samlede vægt deraf, og hvor den første polymer af den kontinuerlige fase ikke er blandbar med den anden polymer af den dispergerede fase, og

(ii) mindst et uorganisk fyldstof, og

(iii) mindst et blødgøringsmiddel,

kendetegnet ved, at

blødgøringsmidlet omfatter diglycerin eller blandinger med diglycerin, og det uorganiske fyldstof omfatter phosphater af calcium og/eller magnesium og omfatter et indhold fra 0,1 til 40 vægtprocent i forhold til den samlede vægt af sammensætningen, og hvor sammensætningen er biologisk nedbrydelig og/eller komposterbar.

2. Termoplastisk sammensætning ifølge det foregående krav, **kendetegnet ved, at**

blødgøringsmidlet omfatter en blanding af:

(a) diglycerin og triglycerin eller

(b) diglycerin og glycerin eller

(c) diglycerin, triglycerin og glycerin,

idet blandingen under (a), (b) og (c) i denne sammensætning omfatter mindst mellem 25 og 75 vægtprocent diglycerin i forhold til summen af (a) diglycerinen og triglycerinen, (b) diglycerinen og glycerinen eller (c) diglycerinen, glycerinen og triglycerinen.

3. Termoplastisk sammensætning ifølge et af de foregående krav,
kendetegnet ved, at
det uorganiske fyldstof har hydrofile egenskaber.

4. Termoplastisk sammensætning ifølge krav 3, **kendetegnet ved, at** calciumphosphaterne er fortrinsvis apatit eller salte og kondensater af orthophosphorsyre, og magnesiumphosphaterne er monobasiske, dibasiske og tribasiske magnesiumphosphater og hydraterne deraf.

5. Termoplastisk sammensætning ifølge et af de foregående krav, **kendetegnet ved, at**
den første polymer af den kontinuerlige fase
- (a) omfatter biogene eller delvist biogene komponenter og/eller er valgt fra en gruppe af polyhydroxysyrer, polyhydroxyalkanoater, polylactider og ligniner, såvel som derivater deraf og/eller copolymerer og især er valgt fra en gruppen bestående af polybutylen-adipat-terephthalat (PBAT), polybutylensebacinat-terephthalat (PBST), polybutylensuccinat (PBS), polybutylensuccinat-adipat (PBSA), og/eller
- (b) omfatter syntetiske komponenter og er valgt fra en gruppe af alifatiske og aromatiske polyestere såvel som alifatiske-aromatiske copolyestere, for eksempel polybutylen-adipat-terephthalat (PBAT).

6. Termoplastisk sammensætning ifølge et af de foregående krav, **kendetegnet ved, at**
sammensætningen som en yderligere dispergeret fase omfatter mindst en amorf eller delvist krystallinsk og især biogen polymer.

7. Termoplastisk sammensætning ifølge krav 6, **kendetegnet ved, at**
den amorfe eller delvist krystallinske og især biogene polymer er en polymer eller copolymer af polymælkesyre (PLA) eller en polymer eller copolymer af polyhydroxyalkanoat (PHA) eller en blanding af mindst to af de førnævnte polymerer.

8. Termoplastisk sammensætning ifølge et af de foregående krav,
kendetegnet ved, at

det biogene tværbindingsmiddel af den dispergerede fase omfatter polysaccharidstrukturelementer, som stammer fra en gruppe, der omfatter stivelser, som hver især indeholder en specifik sammensætning af amylose og amylopektin, såsom for eksempel af kartofler, majs, boghvede, hvede, rug, havre, byg, ris, ærter og tapioka.

9. Termoplastisk sammensætning ifølge krav 8, **kendetegnet ved, at** det biogene tværbindingsmiddel af den dispergerede fase omfatter mindst et yderligere polysaccharidstrukturelement, især valgt fra gruppen bestående af cellulose, hemicellulose og/eller derivater deraf, især acetat-, ester- og etherderivater.

10. Termoplastisk sammensætning ifølge krav 6, **kendetegnet ved, at** forholdet mellem polysaccharidstrukturelementet af den dispergerede fase og den mindst ene første polymer af den kontinuerlige fase er 10 til 90 vægtprocent til 90 til 10 vægtprocent, fortrinsvis i et forhold fra 30 til 70 vægtprocent til 70 til 30 vægtprocent og mere fortrinsvis i et forhold fra 60 til 40 vægtprocent til 40 til 60 vægtprocent i forhold til summen af polysaccharidstrukturelementet af den dispergerede fase og den mindst ene første polymer af den kontinuerlige fase.

11. Fremgangsmåde til fremstilling af folier, film, formstøbte legemer, skummaterialer, belægninger, lakker og dispersioner under anvendelse af den termoplastiske sammensætning ifølge et af kravene 1 til 10.

12. Anvendelse af en termoplastisk sammensætning ifølge et af kravene 1 til 11 til fremstilling af folier, film, formstøbte legemer, skummaterialer, belægninger, lakker og dispersioner ifølge krav 11.

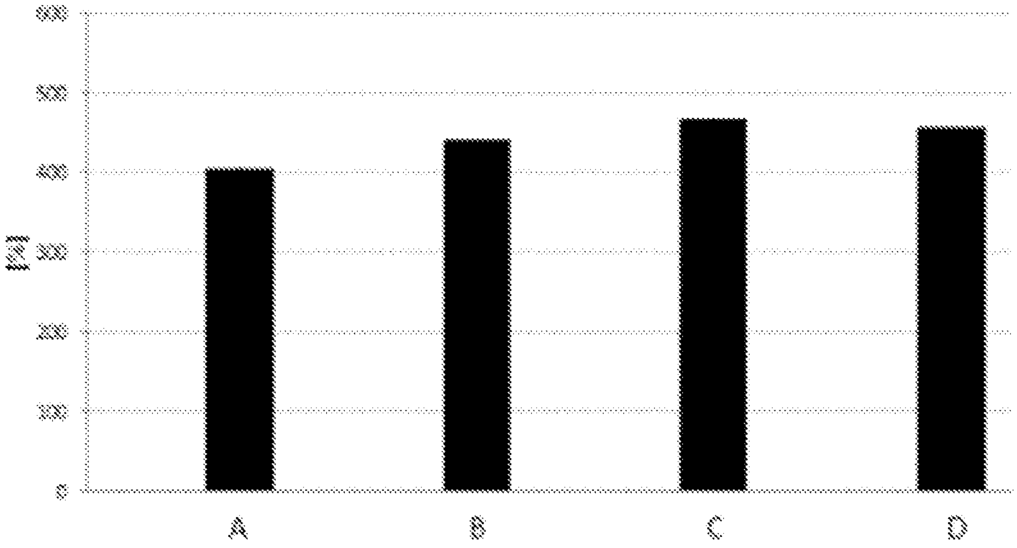

Fig. 1

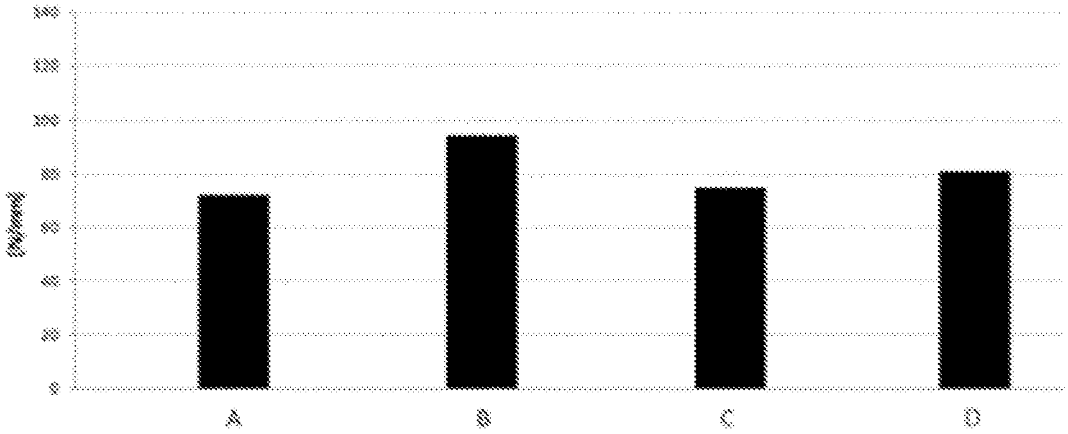

Fig.2

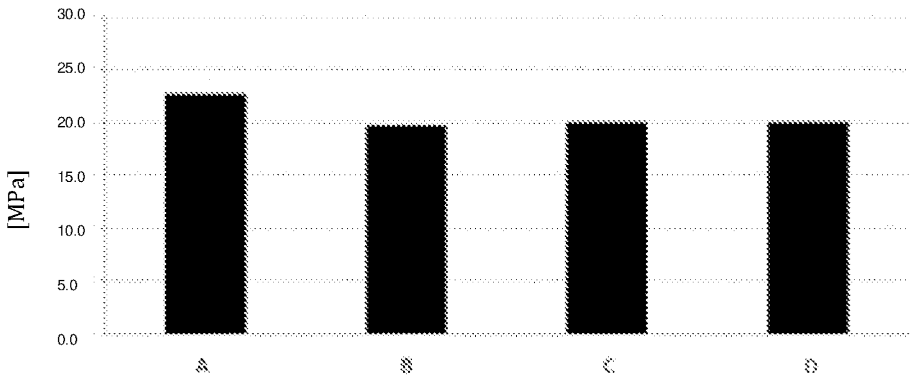
[MPa]
30.0
25.0
20.0
15.0
10.0
5.0
0.0
A
B
C
D

Fig. 3

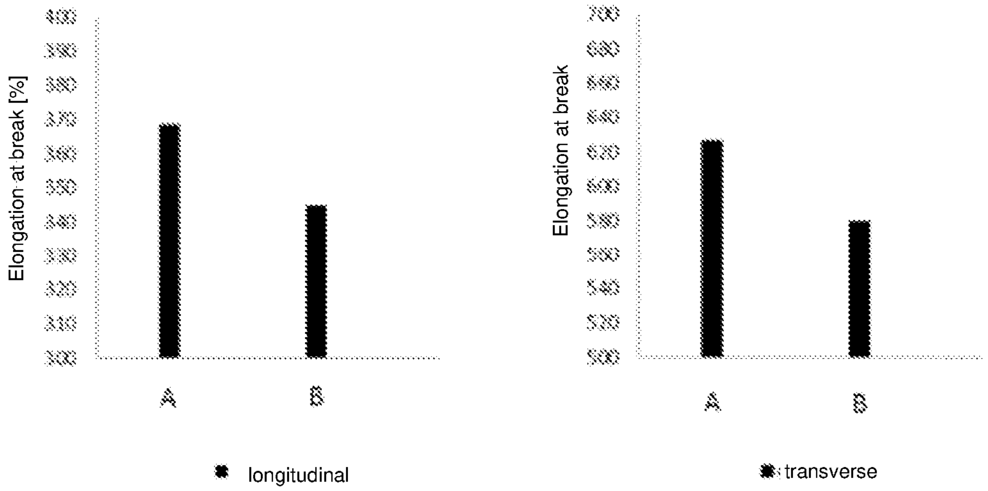
Elongation at break [%]
400
390
380
370
360
350
340
330
320
310
300
A
B
● longitudinal
Elongation at break
700
680
660
640
620
600
580
560
540
520
500
A
B
● transverse

Fig. 4

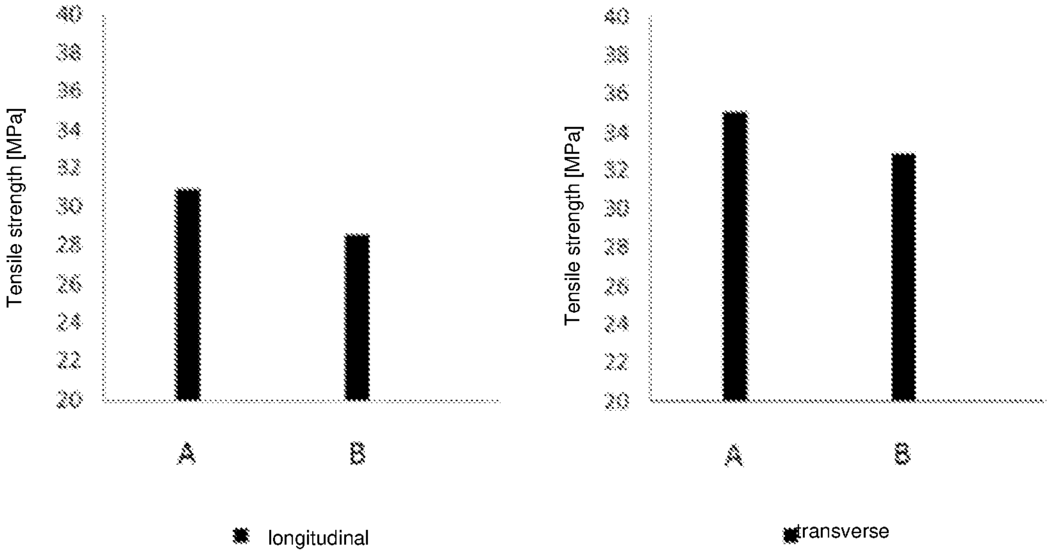
Tensile strength [MPa]
40
38
36
34
32
30
28
26
24
22
20
A
B
longitudinal
Tensile strength [MPa]
40
38
36
34
32
30
28
26
24
22
20
A
B
transverse

Fig. 5

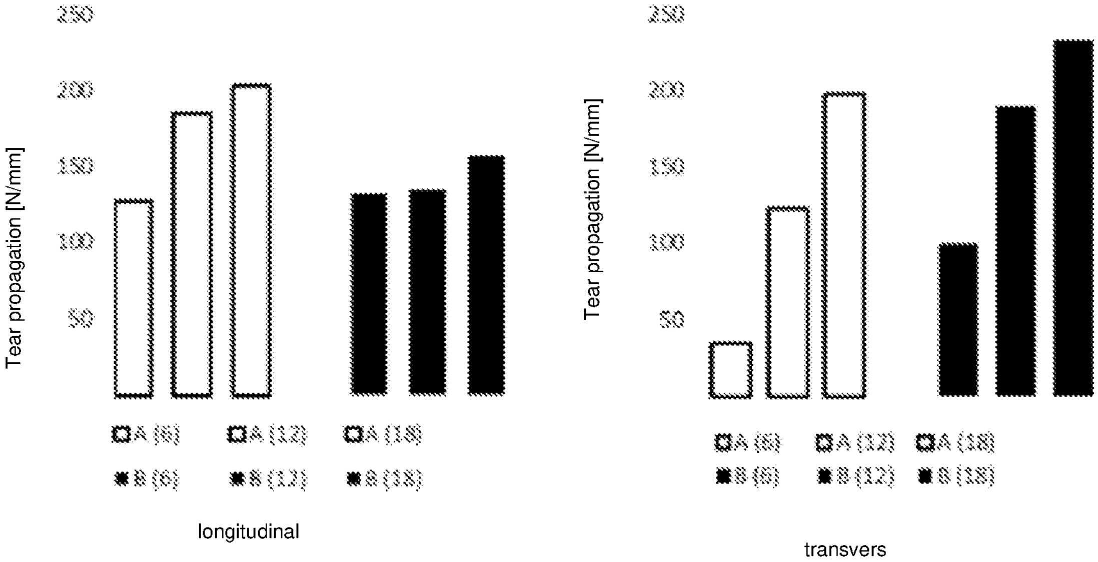
Tear propagation [N/mm]
250
200
150
100
50
A (6)
A (12)
A (18)
B (6)
B (12)
B (18)
longitudinal
Tear propagation [N/mm]
250
200
150
100
50
A (6)
A (12)
A (18)
B (6)
B (12)
B (18)
transvers

Fig. 6

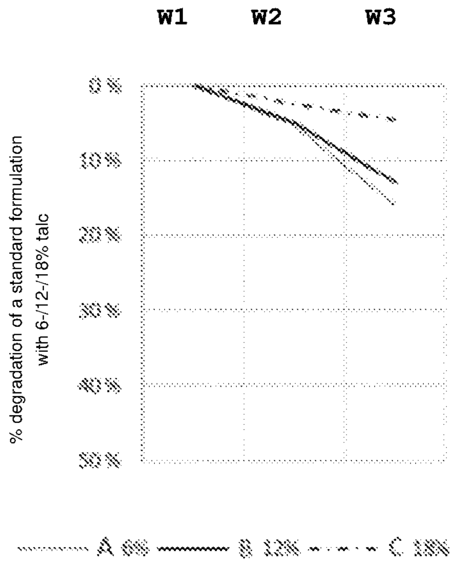
W1
W2
W3
% degradation of a standard formulation with 6-/12-/18% talc
0 %
10 %
20 %
30 %
40 %
50 %
A 6%
B 12%
C 18%

Fig. 7

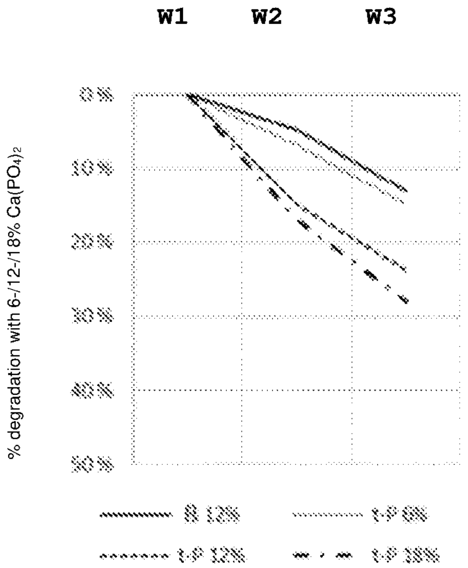
W1
W2
W3
% degradation with 6-/12-/18% Ca(PO4)2
0 %
10 %
20 %
30 %
40 %
50 %
B 12%
t-P 6%
t-P 12%
t-P 18%

Fig. 8

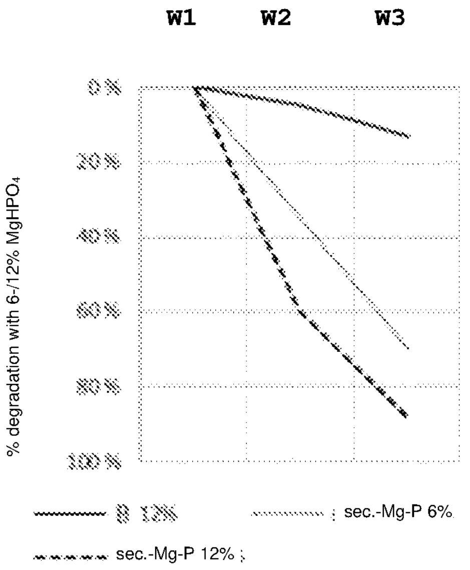
W1
W2
W3
% degradation with 6-/12% MgHPO₄
0 %
20 %
40 %
60 %
80 %
100 %
B 12%
; sec.-Mg-P 6%
sec.-Mg-P 12% ;

Fig. 9