

(10) International Publication Number WO 2011/110750 A1

(43) International Publication Date 15 September 2011 (15.09.2011)

(51) International Patent Classification: C09D 167/04 (2006.01) **B65D 65/46** (2006.01) B32B 27/12 (2006.01) C08L 67/04 (2006.01)

B32B 27/36 (2006.01)

(21) International Application Number:

PCT/FI2011/050215

(22) International Filing Date:

11 March 2011 (11.03.2011)

(25) Filing Language:

Finnish

(26) Publication Language:

English

(30) Priority Data:

20105247 12 March 2010 (12.03.2010)

(71) Applicant (for all designated States except US): STORA

- (72) Inventors; and
- (75) Inventors/Applicants (for US only): PENTTINEN, Tapani [FI/FI]; Lökörentie 31, FI-49210 Huutjärvi (FI). NEVALAINEN, Kimmo [FI/FI]; Raamikatu 13, FI-48910 Kotka (FI). KUUSIPALO, Jurkka [FI/FI]; Sammonkatu 34 G 84, FI-33540 Tampere (FI). KOSKI-NEN, Tapio [FI/FI]; Atanväylä 13 D 14, FI-33580 Tampere (FI). KOTKAMO, Sami [FI/FI]; Pohjolankatu 6-8 C 41, FI-33500 Tampere (FI).

ENSO OYJ [FI/FI]; PL 309, FI-00101 Helsinki (FI).

- (74) Agent: BERGGREN OY AB; P.O. Box 16, (Antinkatu 3 C), FI-00101 Helsinki (FI).
- (81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PE, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.
- (84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

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

(54) Title: HEAT-SEALABLE BIODEGRADABLE PACKAGING MATERIAL, A METHOD FOR ITS MANUFACTURE, AND A PRODUCT PACKAGE MADE FROM THE MATERIAL

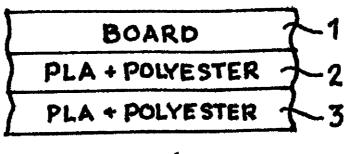


Fig. 7

(57) Abstract: The invention relates to a heat-sealable biodegradable packaging material, its manufacturing method and a product package formed thereof. The packaging material comprises a fibre substrate (1), an inner polymeric coating layer (2) against the same, which contains polylactide and a biodegradable polyester that is blended therewith to improve the adhesion between the layer and the fibre substrate, and an outer coating layer (3) that constitutes the outer surface of the material and contains polylactide and a biodegradable polyester that is blended therewith to improve the heat sealing ability of the layer, the portion of polylactide in the inner layer (2) being higher than that in the outer layer (3). The inner and/or outer coating layers (2, 3) can further have blended therewith a small amount of acrylic copolymer that improves the adhesion and/or heat sealing ability. The coating layers can be introduced onto the fibre substrate by extrusion or coextrusion.



5

10

25

Heat-sealable biodegradable packaging material, a method for its manufacture, and a product package made from the material

The invention relates to a heat-sealable, biodegradable packaging material comprising a fibre substrate and polymeric coating layers extruded onto the same. The invention further relates to the manufacturing method of such packaging material and a closed product package made from the material.

The fibre-based packaging material of product packages, such as packing paper or board, is usually provided with a polymeric coating that makes the package tight and by means of which the package can be closed by heat sealing. Multi-layer coatings can comprise an inner EVOH, PET or polyamide layer that provides the material with an effective barrier to water vapour and oxygen, and an outer polyolefin layer that makes the material heat-sealable. One disadvantage of the said widely-used coating polymers is, however, that they are not biodegradable.

Polylactide (PLA), which has reasonably good moisture and gas barrier properties that are adequate to many applications, has been used as the coating polymer of biodegradable packaging material; however, its use involves a number of problems. Polylactide as such is stiff and fragile, requiring a high extrusion temperature and a fairly large layer thickness to stick to the fibre substrate of the packaging material. Because of the high temperature, polylactide runs the risk of breaking, and in extrusion, the edges of a molten web tend to tear and pin holes easily remain in the extruded layer.

As a solution to said problems, the specification FI-112624 B (EP-1094944 B1, respectively) discloses an inner adhesion layer, which is co-extruded together with an outer polylactide layer and which consists of a biodegradable polymer, examples of which, according to the specification, include some commercial copolyesters, cellulose esters, and polyester amides. They facilitate the extrusion of polylactide and provide adhesion that prevents the coating from peeling off the fibre substrate.

Another problem with the use of polylactide in the outer coating layer of the packaging material is its fairly high melting point, and the resulting poor heat sealing ability. As an improvement to this, the specification US-2002/0065345 A1 discloses a biodegradable aliphatic polyester that is blended with polylactide, its portion in the mixture being at least 9 %, and a tackifier, its portion in the mixture being at least 1 %. As suitable aliphatic polyesters, the publication mentions polycaprolactone (PLC) and polybutylene succinate adipate (PBSA). According to the patent specification, the mixture can be extruded into a film, which can be axially or biaxially stretched and which can be attached to the fibre substrate by lamination. As a result, polymer-coated biodegradable packaging material is obtained, which has a considerably improved heat sealing ability.

5

10

15

20

25

30

35

The specification US 2005/0192410 A1 describes polylactide films and coatings, wherein the processibility of polylactide is improved by blending with it 10–40 weight-% of polycaprolactone and 5–10 weight-% of mineral particles. According to the specification, the mixture can be used in extrusion coating, but there is no reference to its adhesion to the fibre substrate or to its heat sealing ability in the specification. Instead, the specification describes the intermediate layers between the carrier and the PLA-based coating layer or the top layers that come on top of the PLA layer; cf. section [0039] in the specification.

The specification US 2007/0259195 A1 describes polylactide-based films, which contain, blended therewith, 0.1–10 weight-% of biodegradable polymeric additive, the purpose of which is to increase the crystallinity of the polylactide, improving its heat resistance. As examples of such additives, the specification presents FEPOL 2040 marketed by Far Eastern Textile, Taiwan, and Ecoflex marketed by BASF, both of which comprise polybutylene adipate terephthalate (PBAT). According to the specification, the mixtures can be extruded onto the fibre substrate in a conventional manner, but there is no reference to the adhesion of the mixture to the substrate or to the heat sealing ability of the coating thus obtained. In the specification, the intended improved heat resistance of PLA does not, however, refer to an improvement of the heat sealing ability but rather to its weakening.

The purpose of the present invention is to provide polymer-coated, biodegradable packaging material, wherein the coating produced by extrusion and containing polylactide has both an improved adhesion to the fibre substrate and improved heat sealing ability. According to the invention, the solution is that the material includes an inner coating layer against the fibre substrate, containing polylactide and a biodegradable polyester blended therewith to improve the adhesion between the layer and the fibre substrate, and an outer coating layer that forms the outer surface of the material and contains polylactide and a biodegradable polyester blended therewith to improve the heat sealing ability of the layer, the portion of polylactide in the inner layer being higher than that in the outer layer.

3

In the invention, the biodegradable polyester that is blended with polylactide considerably improves the adhesion of polylactide, which by definition is weak, when it is extruded onto the fibre substrate, as well as the heat sealing ability of the extruded polylactide coating to an uncoated fibre substrate, in particular, but also to another film or coating layer containing polylactide. Consequently, the invention provides an improved heat sealing ability of the coating without requiring the multistage film lamination according to the specification US 2002/0065345 A1 for producing the coating.

5

10

15

20

25

30

35

According to the invention, it has further been observed that, even though the added polyester improves both the adhesion of polylactide to the fibre substrate in the extrusion and its heat sealing ability, a smaller amount of added polyester is sufficient to improve the adhesion than to improve the heat sealing ability. Accordingly, optimal properties are achieved by dividing the coatings into two layers, of which the inner adhesion layer contains relatively more polylactide than the outer heat sealing layer. The solution also provides savings in material costs, taking into consideration that the biodegradable polyesters that are blended are considerably more expensive than polylactide. In the inner adhesion layer and the outer heat sealing layer of the material, the same biodegradable polyester can be used as an admixture. Polyester in the invention refers to a polymer formed by at least one diol and at least one dicarboxylic acid or similar dihydric organic acid. Potential polyesters include aliphatic polyesters, such as PCL, for example, Mater-Bi marketed by Novamont, PBSA, polybutylene succinate (PBS), such as GsPLA marketed by Mitsubishi, or an aliphatic-aromatic copolyester, such as PBAT, which is a copolymer polymerized from 1,4-butanediol, adipic acid and terephthalic acid monomers. An especially advantageous PBAT material is Ecoflex® marketed by BASF.

According to a preferred embodiment of the invention, the adhesiveness of the inner coating layer that lies against the fibre substrate and/or the heat sealing ability of the outer layer that constitutes the outer surface of the material are further improved by a small amount (suitably 0.5–5 weight-%) of added acrylic copolymer. A preferable additive is, for example, ethylene butyl acrylate glycidyl methacrylate terpolymer. When the content of acrylic polymer in the coating layer is 5 weight-% at the most, it does not prevent the biodegradation of the packaging material in composting or in a dumping ground, even if the acrylic copolymer itself is not biodegradable.

4

According to the invention, the inner coating layer (adhesion layer) located against the fibre substrate can contain 55–95 weight-% of polylactide, 5–40 weight-% of biodegradable polyester, and 0–5 weight-% of acrylic copolymer. The portions of the said components are preferably within 65–90 weight-% of polylactide, 10–30 weight-% of biodegradable polyester, and 0–5 weight-% of acrylic copolymer.

5

10

15

20

25

30

35

The outer heat-sealable coating layer (heat sealing layer), in turn, can contain 35–90 weight-% of polylactide, 10–60 weight-% of biodegradable polyester, and 0–5 weight-% of acrylic copolymer. The portions of the components are preferably within 50–80 weight-% of polylactide, 20–45 weight-% of biodegradable polyester, and 0–5 weight-% of acrylic copolymer.

The packaging material can further be varied within the invention, so that one or more biodegradable polymeric intermediate layers are placed between the said inner adhesion layer and the outer heat sealing layer to improve the technical properties of the multi-layer coating and/or because of costs. The intermediate layer can comprise, for example, mainly mere polylactide, whereby savings are made in the use of the more expensive polyester by delimiting it to only the thinner adhesion and heat sealing layers on both sides of the thicker polylactide layer. By thickening the coating by means of the PLA layer, the water vapour and oxygen barrier formed by it is improved. In addition to or instead of this, polymer layers that form a more effective water vapour or oxygen barrier can be arranged in the material, for example, from polyvinyl alcohol (PVOH) or polyglycolic acid (PGA).

The method according to the invention for manufacturing the packaging material described above is characterized in that at least two superimposed polymeric coating layers are extruded onto the fibre substrate, of which the inner coating layer that comes against the fibre substrate contains polylactide and a biodegradable polyester that is blended therewith to improve the adhesion between the layer and the fibre substrate, and the outer coating layer that constitutes the outer surface of the material contains polylactide and a biodegradable polyester that is blended therewith to improve the heat sealing ability of the layer, the portion of polylactide in the inner layer being higher than that in the outer layer.

As already mentioned, biodegradable intermediate layers that simply consist, for example, of polylactide and/or said biodegradable barrier polymers, such as PGA, can be incorporated into the coating between the inner adhesion layer and the outer heat sealing layer. All superimposed coating layers can be brought onto the fibre substrate by coextrusion at one stage.

The closed package formed from the packaging material described above, according to the invention, is characterized in that it is closed by heat sealing the coating layer, which contains polylactide and the biodegradable polyester blended therewith and which forms the surface of the material. The heat sealing layer is particularly advantageously sealed to uncoated fibre substrate on the opposite side of the material. Instead of such packaging material that is coated on one side only, material that is coated on both sides can also be used, wherein the package is closed by sealing the coating layers to one another.

5

10

15

20

25

30

35

In the following, the invention is described in more detail by means of examples and with reference to the appended drawing, wherein figs. 1–8 show the sheet constructions of various embodiments of the packaging material according to the invention.

The embodiment of the invention according to fig. 1 comprises a fibre substrate, such as packing paper or board 1, and two polymeric coating layers 2, 3 that are co-extruded onto the same. The inner coating layer 2 consists of polylactide (PLA) and biodegradable polyester that is blended therewith, the purpose of which is to improve the adhesion between the layer 2 and the fibre substrate 1. The outer coating layer 3 also consists of polylactide and biodegradable polyester, the purpose of which is to improve the heat sealing ability of the layer 3. In particular, the outer layer 3 can be heat-sealed to the uncoated surface of the fibre substrate, such as the packing board 1, i.e., on the counter surface of the material according to fig. 1, or to a corresponding heat-sealable polymer surface, for example, when folding the material into a bag and seaming it to itself. The polyester that is blended into the coating layers 2, 3 can comprise the same polyester, or different polyesters can be used in the layer. It is essential for the invention that the portion of polylactide in the inner coating layer 2 is higher than that in the outer coating layer 3.

Fig. 2 shows the same packaging material sheet construction as fig. 1, but the polyester included in the coating layers 2, 3 herein is defined, in an exemplary manner, to comprise the copolyester (PBAT) that is polymerized from 1,4-butanediol, adipic acid and terephthalic acid monomers. In the inner coating layer 2 lying against the fibre substrate 1, the portion of polylactide can be, for example, 60–95, preferably 70–90 weight-%, and the portion of PBAT, respectively, 5–40, preferably 10–30 weight-%. In the outer heat-sealable coating layer 3, the portion of polylactide can be 40–90, preferably 55–80 weight-%, and that of PBAT, re-

5

10

15

20

25

30

spectively, 10–60, preferably 20–45 weight-%. It is required, however, that the portion of polylactide in the inner layer 2 is higher than in the outer layer 3.

The weight of the fibre substrate 1 in the embodiment according to figs. 1 and 2 can be within $40-350 \text{ g/m}^2$; the weight of the inner coating layer within 5-20, preferably $5-10 \text{ g/m}^2$, and the weight of the outer layer within 5-20, preferably $5-10 \text{ g/m}^2$.

In the embodiment according to fig. 3, in the construction according to fig. 1, an adhesive coating layer 2 is added on the opposite side of the fibre substrate 1. In terms of its composition and layer weight, this layer 2 corresponds to that which was presented above in regard to the adhesive layer 2 of figs. 1 and 2. When forming the package, the heat sealing layer 3 can be sealed to the adhesive coating layer 2 of the opposite side of the paper board.

In fig. 4, the fibre substrate 1 is provided on both sides thereof with polymeric coating layers 2, 3 that correspond to fig. 1, so that the coated paper board has a symmetrical construction. When forming the package, the heat sealing layers 3 of the opposite sides of the fibre substrate 1 can be sealed to one another.

The packaging materials according to figs. 5 and 6 differ from the corresponding materials according to figs. 1 and 2 only in that a layer of polylactide 4 is placed between the inner, adhesive coating layer 2 and the outer, heat sealing coating layer 3, its layer weight being 5–40, preferably 10–30 g/m². That which is presented above in connection with figs. 1 and 2 applies to the weights of the other layers 1, 2, and 4 of the construction.

Fig. 7 shows an application of the constructions according to figs. 1 and 2, wherein both the inner and outer coating layers 2, 3 comprise, in addition to polylactide and polyester, also a blended acrylic copolymer, which in the figure is specified so as to be ethylene butylene acrylate glycidyl methacrylate terpolymer, in an exemplary manner. The portion of acrylic copolymer in each layer 2, 3 can be within 0.5–5 weight-%. In the inner layer 2, the acrylic copolymer has the effect of improving the adhesion between the layer and the fibre substrate 1, and in the outer layer 3, the heat sealing ability of the layer. As an advantageous specific composition for the polymer of the inner layer 2, the following could be suggested: 85 weight-% of polylactide, 10 weight-% of 1,4-butanediol/adipic acid/terephthalic acid copolyester (PBAT), and 5 weight-% of ethylene butylene acrylate glycidyl methacrylate terpolymer, and for the polymer of the outer layer 3: 70 weight-% of polylactide, 25

weight-% of PBAT, and 5 weight-% of ethylene butylene acrylate glycidyl methacrylate terpolymer. Suitable layer weights in the construction of fig. 7 are the same as those in the constructions of figs. 1 and 2.

The embodiment according to fig. 8 differs from the one in fig. 7 only in that a layer of PLA 4 is placed between the inner adhesive coating layer 2 and the outer heat sealing coating layer 3, in a similar manner as in figs. 3 and 4. Suitable layer weights for each layer 1–4 correspond to those described above.

Fig. 9 shows a development of the sheet construction according to figs. 5 and 6, where instead of the intermediate layer 4 of polylactide, there are a layer of polyglycolic acid (PGA) 5 and layers of polylactide 6, 7 on both sides of the PGA layer 5. The fibre substrate 1 thus has a polymeric five-layer coating, which is introduced onto the fibre substrate by coextrusion, similarly to the multi-layer coatings according to the figs. 1–6 above. In the construction of fig. 9, PGA can be replaced by polyvinyl alcohol (PVOH).

15 Examples

5

10

25

30

1. Adhesion

Various polymeric single-layer coatings were introduced onto the paper board by extrusion, and their adhesion to the board surface was defined on a scale of 1–5, whereby the classification was as follows:

- 1 = no adhesion, the polymeric layer peels off;
- 2 = poor adhesion, some fibres are stuck to the polymeric layer that peels off;
- 3 = poor adhesion, when detaching the polymeric layer, less than 50% of the paper board breaks in the area of coating;
 - 4 = moderate adhesion, when detaching the polymeric layer, over 50% of the paper board breaks in the area of coating;
 - 5 = perfect adhesion, when detaching the polymeric layer, the paper board breaks throughout the area of coating.

The coating polymers tested comprised polylactide (PLA) as such or blended with various polyesters (polyesters 1–6) or with polyesters and an additive that consisted of acrylic copolymer. The polyesters and the additive were:

Polyester 1 = PBS

35 Polyester 2 = PBS

Polyester 3 = PBAT

Polyester 4 = PBAT

Polyester 5 = PBSA

Polyester 6 = PBAT

Additive = ethylene butylene acrylate glycidyl methacrylate terpolymer.

5

The results for the various polymer compositions and layer weights are in the following table 1. Table 2 further shows results for various polymer compositions on a layer weight of 25 g/m². In each polymer mixture, PLA constitutes the rest of the mixture, so that the portions of components total in 100%.

10 **Table 1**

Polymer	Layer weight (g/m²)	Adhesion
PLA	38.8	5
PLA	31.1	5
PLA	25.7	4
PLA	20.7	4
PLA	18.9	3
PLA+ 50% Polyester 1	18.6	5
PLA+ 25% Polyester 1	19.0	5
PLA+ 25% Polyester 1	16.7	5
PLA+ 25% Polyester 1	14.1	4
PLA+ 10% Polyester 2	19.3	5
PLA+ 10% Polyester 2	12.7	3
PLA+ 10% Polyester 2	6.0	1
PLA+ 40% Polyester 3	16.4	5
PLA+ 40% Polyester 3	14.06	4
PLA+ 20% Polyester 3	22.3	5
PLA+ 20% Polyester 3	15.5	4
PLA+ 20% Polyester 3	12.8	3
PLA+ 40% Polyester 4	15.72	5
PLA+ 40% Polyester 4	12.3	3
PLA+ 40% Polyester 4	8.42	2
PLA+ 50% Polyester 5	24.26	5
PLA+ 50% Polyester 5	19.18	5
PLA+ 30% Polyester 5	21.5	5
PLA+ 30% Polyester 5	20.78	4
PLA+ 30% Polyester 5	15.9	4
Polymer	Layer weight (g/m²)	Adhesion
PLA+ 30% Polyester 5	15.64	4
PLA+ 30% Polyester 5	12.28	3
PLA+ 25% Polyester 5	26.68	5
PLA+ 25% Polyester 5	20.72	5
PLA+ 25% Polyester 5	17.24	4
PLA+ 20% Polyester 5	27.58	5
PLA+ 20% Polyester 5	22.12	4
PLA+ 20% Polyester 5	19.6	4
PLA+ 10% Polyester 5	32.38	5
PLA+ 10% Polyester 5	26.8	5
PLA+ 10% Polyester 5	21.9	5
PLA+ 40% Polyester 6	24.92	5
PLA+ 40% Polyester 6	23.76	5

Table 1, cont.

Polymer	Layer weight (g/m²)	Adhesion
PLA+ 40% Polyester 6	23.26	4.5
PLA+ 40% Polyester 6	14.5	3.5
PLA+ 20% Polyester 2 + 5% additive	14.2	5
PLA+ 20% Polyester 2 + 5% additive	13.38	4.5
PLA+ 20% Polyester 2 + 5% additive	11.38	3.5
PLA+ 10% Polyester 2 + 5% additive	16.46	5
PLA+ 10% Polyester 2 + 5% additive	14.3	5
PLA+ 10% Polyester 2 + 5% additive	15.66	4.5
PLA+ 10% Polyester 2 + 5% additive	10.72	3
PLA+ 10% Polyester 6 + 5% additive	21.74	5
PLA+ 10% Polyester 6 + 5% additive	19	4.5
PLA+ 10% Polyester 6 + 5% additive	17.26	5
PLA+ 10% Polyester 6 + 5% additive	13.88	3.5
PLA+ 10% Polyester 6 + 5% additive	9.6	2.5
PLA+ 10% Polyester 6 + 5% additive	7.2	2

Table 2

Polymer	Adhesion on a layer weight of 25 g/m ²
PLA	4
PLA+ 50% Polyester 1	5
PLA+ 25% Polyester 1	5
PLA+ 40% Polyester 2	5
PLA+ 10% Polyester 2	5
PLA+ 40% Polyester 3	5
PLA+ 20% Polyester 3	5
PLA+ 10% Polyester 3	5
PLA+ 40% Polyester 4	5
PLA+ 50% Polyester 5	5
PLA+ 30% Polyester 5	5
Polymer	Adhesion on a layer weight of 25
	g/m²
PLA+ 25% Polyester 5	5
PLA+ 20% Polyester 5	5
PLA+ 10% Polyester 5	5
PLA+ 40% Polyester 6	5
PLA+ 20% Polyester 2 + 5% additive	5
PLA+ 10% Polyester 2 + 5% additive	5
PLA+ 10% Polyester 6 + 5% additive	5

5

10

The results indicate that the added polyesters improve the adhesion of polylactide, which is indicated by achieving a perfect adhesion on lower coating layer weights. Generally speaking, when the portion of polyester in the mixture increases, the adhesion improves, but when using a practicable layer weight of 25 g/m², acceptable adhesion can be achieved with a mixture portion as low as 10–20%. It is also obvious that the added acrylic copolymer further improves the adhesion.

10

2. Heat sealing onto the paper board surface

Pieces of paper board that were extrusion-coated with different polymers were heat-sealed onto a blank board surface. Sealing between the polymer coating and the board surface was defined on a scale of 1–5, whereby the classification was:

- 5 1 = no sealing
 - 2 = poor sealing, sealed surfaces could be detached from each other essentially in one piece;
 - 3 = poor sealing, when detached, less than 50% of the board breaks in the area of polymer coating;
- 4 = moderate sealing, when detached, over 50% of the board breaks in the area of polymer coating;
 - 5 = perfect sealing, when detached, the board breaks throughout the area of coating.
- The coating polymers consisted of polylactide or mixtures of polylactide and polyesters and possibly the acrylic polymer that was added as an additive, as above in
 the adhesion testing. The results, in other words, sealing at different sealing temperatures, are shown in table 3. Furthermore, the table 4 includes the lowest sealing temperature of each coating polymer for achieving a perfect sealing and an
 evaluation of the sealing ability based on the same.

Table 3

Polymer	Heat sealing temperature (°C)												
	100	110	120	125	130	135	140	145	150	155	160	165	170
PLA	1	2	2	3	3	3	3	3.5	4.5	4.5	4.5	4.5	5
PLA+ 40% Polyester 3		2	2.67	2.67	3	3	3.67	4.33	5	5	5	5	5
PLA+ 50% Polyester 5	3	3.33	4.33	5	5	5	5	5	5	5	5	5	5
PLA+ 25% Polyester 5	2		3	3	3.67	4	4.33	5	5	5	5	5	5
PLA+ 40% Polyester 6			3		3.5	3.5	4.5	4.5	5	5	5	5	5
PLA+ 10% Polyester 6 + 5% additive	1	2	3	3	3.5	3.5	4.5	4.5	5	5	5	5	5
PLA + 5% additive		2	2.33	2.67	3	3	3.33	4	4.67	4.67	5	5	5

Table 4

5

10

Polymer	Lowest sealing temperature	Sealing ability
	(°C)	
PLA	170	Poor
PLA+ 40% Polyester 3	150	Good
PLA+ 50% Polyester 5	125	Excellent
PLA+ 25% Polyester 5	145	Good
PLA+ 40% Polyester 6	150	Good
PLA+ 10% Polyester 6 + 5% additive	150	Good
PLA + 5% additive	160	Sufficient

The results indicate that the added polyesters improve the heat sealing ability of polylactide, which is evidenced by achieving a perfect sealability at lower sealing temperatures. Comparison of the results obtained with the different portions of polyester 5 in the mixtures proves that when the portion of polyester increases, the sealing temperature can be decreased, i.e., the sealing ability improves. An acceptable sealing temperature of 150 °C requires an about 25–40% portion of polyester in the mixture. The added acrylic polymer, however, enables a considerable decrease in the amount of polyester.

CLAIMS

5

10

15

35

- 1. Heat-sealable biodegradable packaging material, comprising a fibre substrate (1) and polymeric coating layers (2-8) that are extruded onto the same, characterized in that the material includes an inner coating layer (2) that is placed against the fibre substrate and contains polylactide and a biodegradable polyester that is blended therewith to improve adhesion between the layer and the fibre substrate, and an outer coating layer (3) that constitutes the outer surface of the material and contains polylactide and a biodegradable polyester that is blended therewith to improve the heat sealing ability of the layer, the portion of polylactide in the inner layer (2) being higher than in the outer layer (3).
- 2. Packaging material according to claim 1, **characterized** in that the inner and outer coating layers (2, 3) contain the same biodegradable polyester, blended therewith.
- 3. Packaging material according to claim 1 or 2, **characterized** in that the inner and/or outer coating layers (2, 3) contain a biodegradable aliphatic-aromatic copolyester.
- 20 4. Packaging material according to claim 3, characterized in that the biodegradable copolyester is one that has been polymerized from 1,4-butanediol, adipic acid and terephthalic acid monomers.
- 5. Packaging material according to any of the preceding claims, **characterized** in that the inner and/or outer coating layers (2, 3) further comprise, blended therewith, no more than 5 weight-% of acrylic copolymer that improves the adhesion and/or heat sealing ability.
- 6. Packaging material according to claim 5, **characterized** in that the acrylic copolymer is ethylene butyl acrylate glycidyl methacrylate terpolymer.
 - 7. Packaging material according to any of the preceding claims, **characterized** in that the inner coating layer (2) that is placed against the fibre substrate contains 55–95 weight-% of polylactide, 5–40 weight-% of biodegradable polyester, and 0–5 weight-% of acrylic copolymer.

13

8. Packaging material according to claim 7, **characterized** in that the inner coating layer (2) that is placed against the fibre substrate contains 65–90 weight-% of polylactide, 10–30 weight-% of biodegradable polyester, and 0–5 weight% of acrylic copolymer.

5

9. Packaging material according to any of the preceding claims, **characterized** in that the outer heat-sealable coating layer (3) contains 35–90 weight-% of polylactide, 10–60 weight-% of biodegradable polyester, and 0–5 weight-% of acrylic copolymer.

10

- 10. Packaging material according to claim 9, **characterized** in that the outer heat-sealable coating layer (3) contains 50–80 weight-% of polylactide, 20–45 weight-% of biodegradable polyester, and 0–5 % weight-% of acrylic copolymer.
- 15 11. Packaging material according to any of the preceding claims, **characterized** in that between the said inner and outer coating layers, there is provided at least one polymeric intermediate layer (4–7).
- 12. Packaging material according to claim 11, **characterized** in that an interme-20 diate layer (4, 6, 7) mainly consists of mere polylactide.
 - 13. Packaging material according to claim 11 or 12, **characterized** in that an intermediate layer (5) consists of a biodegradable polymer that forms a barrier to water vapour and/or oxygen, such as polyvinyl alcohol or polyglycolic acid.

25

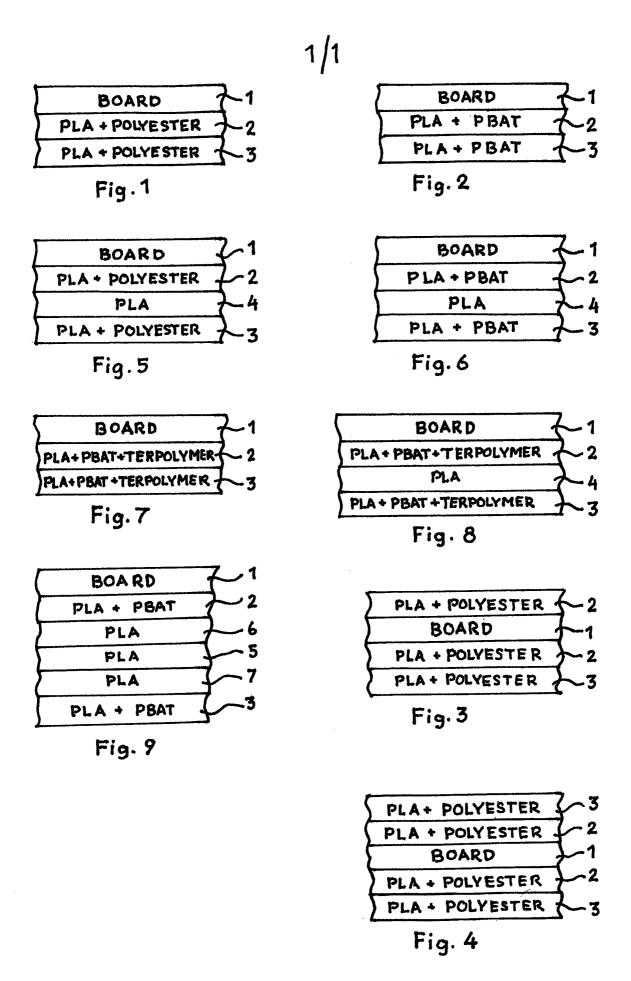
30

14. A method of manufacturing a packaging material according to any of the preceding claims, **characterized** in that at least two superimposed polymeric coating layers are extruded onto the fibre substrate (1), of which the inner coating layer (2) that comes against the fibre substrate contains polylactide and a biodegradable polyester that is blended therewith to improve the adhesion between the layer and the fibre substrate, and the outer coating layer (3) that constitutes the outer surface of the material contains polylactide and a biodegradable polyester that is blended therewith to improve the heat sealing ability of the layer, the portion of polylactide in the inner layer (2) being higher than that in the outer layer (3).

35

15. A method according to claim 14, **characterized** in that at least three superimposed polymeric coating layers are extruded onto the fibre substrate, so that at least one intermediate layer (4, 6, 7) between the said inner and outer coating layers (2, 3) mainly consists of mere polylactide.

- 16. A method according to claim 14 or 15, **characterized** in that at least three superimposed polymeric coating layers are extruded onto the fibre substrate (1), so that at least one intermediate layer (5) between the said inner and outer coating layers (2, 3) consists of a biodegradable polymer, such as polyvinyl alcohol or polyglycolic acid that improves the barrier to water vapour and/or oxygen.
- 17. A method according to any of Claims 14–16, **characterized** in that the coating layers (2–7) are introduced onto the fibre substrate by coextrusion.
- 18. A product package consisting of a packaging material according to any of claims 1–13, which is closed by heat sealing between the top layer (3) containing polylactide and a biodegradable polyester that is blended therewith, and uncoated fibre substrate (1) on the opposite side of the material.



International application No. PCT/FI2011/050215

A. CLASSIFICATION OF SUBJECT MATTER

IPC: see extra sheet

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC: B32B, B65D, C08L, C09D

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

SE, DK, FI, NO classes as above

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

EPO-Internal, PAJ, WPI data

C. DOCUMENTS CONSIDERED TO BE RELEV	ANT
-------------------------------------	-----

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 2009064052 A1 (HANCHANG PAPER CO LTD ET AL), 22 May 2009 (2009-05-22); abstract; page 6, line 11 - line 14; page 14, line 13 - page 17, line 15; claim 1; Table 1	1-18
Y	US 20020065345 A1 (NARITA JUNICHI ET AL), 30 May 2002 (2002-05-30); abstract; [0036], [0041], [0054], [0073]-[0074]	1-18
A	EP 1059162 A2 (TETRA LAVAL HOLDINGS & FINANCE), 13 December 2000 (2000-12-13); abstract; [0058]-[0059]	5-6
А	EP 1504895 A1 (SCHULMAN A PLASTICS), 9 February 2005 (2005-02-09); abstract; claims 1, 16, 19, 28	5-6

Further documents are listed in the continuation of Box C.	See patent family annex.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considere to be of particular relevance	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention		
 "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is 	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone		
cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination		
means "P" document published prior to the international filing date but later than the priority date claimed	being obvious to a person skilled in the art "&" document member of the same patent family		
Date of the actual completion of the international search	Date of mailing of the international search report		
20-06-2011	21-06-2011		
Name and mailing address of the ISA/SE	Authorized officer		
Patent- och registreringsverket Box 5055 S-102 42 STOCKHOLM	Monika Bohlin		
Facsimile No. + 46 8 666 02 86	Telephone No. + 46 8 782 25 00		

International application No.
PCT/FI2011/050215

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5753724 A (EDGINGTON GARRY J ET AL), 19 May 1998 (1998-05-19); abstract	1-18
А	US 6183814 B1 (NANGERONI JAMES ET AL), 6 February 2001 (2001-02-06); abstract; column 4, line 45 - column 7, line 29; column 18, line 1 - line 3; column 28, line 45 - line 62; column 29, line 40 - column 30, line 42	1-18
Α	EP 1094944 A1 (ENSO OYJ), 2 May 2001 (2001-05-02); abstract; figures 2-3	1-18
Α	US 6326440 B1 (TERADA SHIGENORI ET AL), 4 December 2001 (2001-12-04); abstract	1-18
А	EP 1236753 A1 (HYCAIL B V), 4 September 2002 (2002-09-04); abstract	1-18
P, X	JP 2010069767 A (DAINIPPON PRINTING CO LTD), 2 April 2010 (2010-04-02); (abstract) Retrieved from: EPODOC database; Original document: figure 1	1-18

International application No. PCT/FI2011/050215

Continuation of: second sheet

International Patent Classification (IPC)

B65D 65/46 (2006.01)

B32B 27/12 (2006.01)

B32B 27/36 (2006.01)

C09D 167/04 (2006.01)

C08L 67/04 (2006.01)

Download your patent documents at www.prv.se

The cited patent documents can be downloaded:

- From "Cited documents" found under our online services at <u>www.prv.se</u> (English version)
- From "Anförda dokument" found under "e-tjänster" at <u>www.prv.se</u> (Swedish version)

Use the application number as username. The password is YNWLYFQRKC.

Paper copies can be ordered at a cost of 50 SEK per copy from PRV InterPat (telephone number 08-782 28 85).

Cited literature, if any, will be enclosed in paper form.

Information on patent family members

International application No. PCT/FI2011/050215

				•		
WO	2009064052 A1	22/05/2009	KR	100875104	B1	19/12/2008
US	20020065345 A1	30/05/2002	DE	60107358	D1	30/12/2004
			EP	1193294	A2	03/04/2002
			JP	4660035	B2	30/03/2011
			JP	2002173589	Α	21/06/2002
			US	6521336	B2	18/02/2003
EP	1059162 A2	13/12/2000	AT	358575	Т	15/04/2007
			DE	60034175	D1	16/05/2007
			ES	2281318	Т3	01/10/2007
			JP	2001030432	Α	06/02/2001
EP	1504895 A1	09/02/2005	AT	319563	Т	15/03/2006
			CA	2476639	Α1	08/02/2005
			DE	60303844	D1	04/05/2006
			ES	2259744	Т3	16/10/2006
			MX	PA04007551	Α	27/05/2005
			US	20080187744	A 1	07/08/2008
			US	20080103259	Α1	01/05/2008
			US	7754340	B2	13/07/2010
			US	7271220	B2	18/09/2007
			US	20050038192	Α1	17/02/2005
			US	7615174	B2	10/11/2009
US	5753724 A	19/05/1998	US	5700344	Α	23/12/1997
US	6183814 B1	06/02/2001	ΑU	733819	B2	24/05/2001
			ΑU	7586298	Α	11/12/1998
			BR	9809136	Α	04/12/2001
			CA	2291172	С	15/04/2008
			EP	0981666	A 1	01/03/2000
			JP	2002513449	T	08/05/2002
			NO	995734		24/01/2000
			ΝZ	501695		25/07/2003
			WO	9853141	A1	26/11/1998

Information on patent family members

International application No. PCT/FI2011/050215

EP	1094944 A1	02/05/2001	AT	277758	Т	15/10/2004
			AU	5040799	Α	24/01/2000
			AU	750072	B2	11/07/2002
			CA	2336898	С	29/05/2007
			DE	69920703	T2	16/02/2006
			ES	2226410	Т3	16/03/2005
			FI	112624	B1	31/12/2003
			FI	981558	Α	08/01/2000
			JP	2002519222	Τ	02/07/2002
			US	6645584	B1	11/11/2003
			WO	0001530	A 1	13/01/2000
			ZA	200100251	Α	04/10/2002
US	6326440 B1	04/12/2001	CA	2309828	A 1	27/05/1999
			CN	1109060	С	21/05/2003
			CN	1280592	Α	17/01/2001
			DE	69827030	D1	18/11/2004
			EP	1033383	A 1	06/09/2000
			ID	27669	Α	19/04/2001
			TW	396108	В	01/07/2000
			WO	9925758	A1	27/05/1999
EP	1236753 A1	04/09/2002	AT	383386	Т	15/01/2008
			AU	2002236346	A 1	19/09/2002
			DE	60224505	D1	21/02/2008
			DK	1366104	Т3	26/05/2008
			ES	2299555	Т3	01/06/2008
			US	20070243374	Α1	18/10/2007
			US	7868101	B2	11/01/2011
			US	20040143072	Α1	22/07/2004
			US	7465770	B2	16/12/2008
			WO	02070583	A2	12/09/2002
JP	2010069767 A	02/04/2010	NONE			
I						