

BIODEGRADABLE HOT MELT ADHESIVES**CROSS-REFERENCE TO RELATED APPLICATIONS**

[001] This application claims the benefit of the earlier filing date of co-pending provisional application 62/448,483, filed January 20, 2017, the disclosure of which is herein incorporated by reference.

FIELD

[002] The present disclosure relates in general to hot melt adhesives and in particular to hot melt adhesives having improved heat stability, as well as high content of components which are bio-sourced and/ or biodegradable.

BACKGROUND

[003] Hot melt adhesives are used commercially to bond a wide variety of substrates. A principal advantage of hot melt adhesives is their lack of a liquid carrier in the adhesive composition. Without a liquid carrier, there is no need for a drying period after application so that production line speeds can be increased. In situations where organic solvents are used as liquid carriers, their elimination reduces environmental and human risks associated with their use. Elimination of the liquid carrier also reduces the weight and bulk of the adhesive for shipping and storage.

[004] Hot melt adhesives are typically melted and then held molten for a period of time in a heated vessel at a temperature close to the application temperature for the adhesive. The period of time the adhesive is held in a molten state may range from just a few hours to as long as several days. For hot melt adhesives derived from petroleum-based components, this extended heating time poses few problems for the integrity of the adhesive.

[005] Nonetheless, it is still desirable to incorporate biodegradable, recyclable, and/or bio-sourced components into hot melt adhesive compositions due to environmental and health concerns. However, for adhesives derived from components that are bio-sourced and/or biodegradable, the polymeric components that give the adhesive its strength and have been found to be susceptible to the degradation reactions noted above both during the application period and after a bond has formed. As a result, hot melt adhesives derived from bio-sourced components generally exhibit a fairly low heat stability when maintained in a heated, molten state for an extended period of time. Because of this, there have to date been few, if any, commercially success applications of such hot melt adhesives.

[006] It would therefore be advantageous to provide a hot melt adhesive composition which incorporate a high content of bio-sourced and/or biodegradable components, which also exhibits heat stability as compared to earlier hot melt adhesive compositions.

SUMMARY

[006A] In one aspect, the present disclosure refers to a hot melt adhesive comprising:

- from 10 to 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from 1500 to 3000;

- from 40 to 75 weight percent of polylactide having a weight average molecular weight from 10,000 to 18,000;

- from 20 to 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids; and

- from 0.5 to 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms,

wherein all weight percentages are based on the overall weight of the hot melt adhesive.

[006B] In another aspect, the present disclosure refers to an adhesive-coated substrate comprising:

- a substrate having at least a first side; and

- a hot melt adhesive coating applied to at least a portion of the substrate first side, wherein the hot melt adhesive coating comprises

- from 10 to 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from 1500 to 3000,

- from 40 to 75 weight percent of polylactide having a weight average molecular weight from 10,000 to 18,000,

- from 20 to 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids, and

- from .5 to 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms,

wherein all weight percentages are based on the overall weight of the hot melt adhesive.

[006C] In a further aspect, the present disclosure refers to a method for making a hot melt adhesive, comprising the steps of:

melt blending, at a temperature from 140 to 175 °C, a mixture which comprises

- from 10 to 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from 1500 to 3000,
- from 40 to 75 weight percent of polylactide having a weight average molecular weight from 10,000 to 18,000,
- from 20 to 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids, and
- from 0.5 to 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms; and

further blending the mixture with from 1 to 5 weight percent of a carboxylic acid or a carboxylic acid derivative, at a temperature at a temperature from 140 to 175 °C to provide a stabilized hot melt adhesive,

wherein all weight percentages are based on the overall weight of the hot melt adhesive.

[007] The present disclosure provides a hot melt adhesive composition. According to one example, the hot melt adhesive includes at least the following components: (1) from about 10 to about 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from about 1500 to about 3000; (2) from about 40 to about 75 weight percent of polylactide having a weight average molecular weight from about 10,000 to about 18,000; (3) from about 20 to about 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids; and (4) from about 0.5 to about 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms. All of the aforementioned weight percentages are based on the overall weight of the hot melt adhesive composition.

[008] In certain examples of the present disclosure, the hot melt adhesive preferably has a viscosity from about 1000 centipoise to about 6000 centipoise at a

temperature of about 143°C. More preferably, the hot melt adhesive has a viscosity from about 2000 centipoise to about 4000 centipoise at a temperature of about 143°C, and most preferably a viscosity from about 2000 centipoise to about 3000 centipoise at a temperature of about 143°C.

[009] In certain examples of the present disclosure, the lactic acid oligomer or polymer preferably has a viscosity from about 350 centipoise to about 450 centipoise at a temperature of about 280°C.

[010] In some examples of the present disclosure, the polylactide preferably has a viscosity from about 1800 centipoise to about 2200 centipoise at a temperature of about 143°C.

[011] In some examples of the present disclosure, the polylactide also includes a polymer capping group formed by reaction of the polylactide with a carboxylic acid or a carboxylic acid derivative. More preferably, the polymer capping groups is form by

reaction of the polylactide with an acid anhydride. Most preferably, the acid anhydride is propionic anhydride.

[012] In one example of the present disclosure, the polyester preferably has a viscosity from about 15,000 centipoise to about 35,000 centipoise at a temperature of about 216°C. In certain examples of the present disclosure, the polyester also preferably has a weight average molecular weight from about 55,000 to about 72,000.

[013] In some examples of the present disclosure, the polyester also includes a polymer capping group formed by reaction of the polyester with a carboxylic acid or a carboxylic acid derivative. More preferably, the polymer capping groups is form by reaction of the polyester with an acid anhydride. Most preferably, the acid anhydride is propionic anhydride.

[014] In some examples of the present disclosure, the polyester is polybutylene(succinate-co-adipate) ("PBSA"). In certain embodiments of the present disclosure, the one or more dicarboxylic acids in the PBSA preferably include from about 10 to about 30 mole percent adipic acid and from about 70 to about 90 mole percent succinic acid.

[015] In some examples of the present disclosure, the mono-unsaturated short chain fatty acid is preferably crotonic acid.

[016] In another example, the present disclosure provides an adhesive-coated substrate. According to one example, the adhesive-coated substrate includes a substrate having at least a first side; and a hot melt adhesive coating applied to at least of portion of the substrate first side. The hot melt adhesive includes at least the following components: (1) from about 10 to about 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from about 1500 to about 3000; (2) from about 40 to about 75 weight percent of polylactide having a weight average molecular weight from about 10,000 to about 18,000; (3) from about 20 to about 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids; and (4) from about 0.5 to about 5 weight percent of a copolymer of vinyl acetate

and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms. All of the aforementioned weight percentages are based on the overall weight of the hot melt adhesive composition.

[017] In yet another example, the present disclosure provides a method for making a hot melt adhesive. According to one example, the method includes the a first step of melt blending, at a temperature from about 140 to about 175 °C, a mixture which includes at least the following components: (1) from about 10 to about 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from about 1500 to about 3000; (2) from about 40 to about 75 weight percent of polylactide having a weight average molecular weight from about 10,000 to about 18,000; (3) from about 20 to about 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids; and (4) from about 0.5 to about 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms.

[018] The method also includes a second step of further blending the mixture with from about 1 to about 5 weight percent of a carboxylic acid or a carboxylic acid derivative, at a temperature at a temperature from about 140 to about 175 °C to provide a stabilized hot melt adhesive. All of the aforementioned weight percentages are based on the overall weight of the hot melt adhesive.

DETAILED DESCRIPTION

[019] The present disclosure provides a hot melt adhesive composition. According to one embodiment, the hot melt adhesive generally includes at least the following components: (1) from about 10 to about 20 weight percent of a lactic acid oligomer or polymer; (2) from about 40 to about 75 weight percent of polylactide; (3) from about 20 to about 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids; and (4) from about 0.5 to about 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms. All of the aforementioned weight percentages are based on the overall weight of the hot melt adhesive composition.

[020] The first component of the hot melt adhesive composition is a low molecular weight lactic acid oligomer or polymer. In general, the lactic acid oligomer or polymer has a weight average molecular weight from about 1500 to about 3000. The lactic acid oligomer or polymer is generally formed from a lactic acid starting material which enriched in its L- enantiomer relative to its D- enantiomer. Preferably, the lactic acid starting material is at least 97% L-lactic acid.

[021] The lactic acid oligomer or polymer generally makes up from 10 to about 20 weight percent of the hot melt adhesive composition. Preferably, if measured by itself, the lactic acid oligomer or polymer exhibits a viscosity from about 350 centipoise to about 450 centipoise at a temperature of about 280°C. Functionally, within the hot melt adhesive, the lactic acid oligomer or polymer acts as a tackifier, improving the tackiness and wetting of the substrate.

[022] The second component of the hot melt adhesive composition is a polylactide. In general, the polylactide has a weight average molecular weight from about 10,000 to about 18,000. The polylactide generally makes up from 40 to about 75 weight percent of the hot melt adhesive composition. Preferably, if measured by itself, the polylactide exhibits a viscosity from about 1800 centipoise to about 2200 centipoise at a temperature of about 143°C.

[023] In some embodiments of the present disclosure, the polylactide also includes a polymer capping group formed by reaction of the polylactide with a carboxylic acid or a carboxylic acid derivative. More preferably, the polymer capping groups is form by reaction of the polylactide with an acid anhydride. Most preferably, the acid anhydride is propionic anhydride.

[024] The polylactide functions as the base material in the hot melt adhesive. Having a higher molecular weight than the other components and being present in a large weight percentage, the polylactide acts as the backbone of the composition and provides the primary properties of the adhesive.

[025] The third component of the hot melt adhesive composition is a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids. In general, the polyester has a weight average molecular weight from about 55,000 to about 72,000. The polyester generally makes up from 20 to about 35 weight percent of the hot melt adhesive composition. Preferably, if measured by itself, the polyester exhibits a viscosity from about 15,000 centipoise to about 35,000 centipoise at a temperature of about 216°C. Functionally, within the hot melt adhesive, the diol / diacid polymer provides improved plasticity, tensile strength, and heat resistance for the composition.

[026] Suitable diols which may be used for the diol moieties of the polyester include ethylene, propylene, butylene, and hexylene. A combination of multiple diols may also be incorporated into the polyester. Suitable diacids which may be used for the diacid moieties of the polyester include dicarboxylic acids having from 2 to 12 carbon atoms.

[027] In a preferred embodiment according to the present disclosure, the polyester may include moieties formed from butylene, succinic acid, and adipic acid. Thus, the polyester may be polybutylene(succinate-co-adipate) or “PBSA”. In these embodiments, the one or more dicarboxylic acids in the PBSA preferably include from about 10 to about 30 mole percent adipic acid and from about 70 to about 90 mole percent succinic acid.

[028] As with the polylactide, in some instances, the polyester may also include a polymer capping group formed by reaction of the polyester with a carboxylic acid or a carboxylic acid derivative. More preferably, the polymer capping groups is form by reaction of the polyester with an acid anhydride. Most preferably, the acid anhydride is propionic anhydride.

[029] The fourth component of the hot melt adhesive composition is a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms. In general, this copolymer makes up from about 0.5 to about 5 weight percent of the hot melt adhesive composition. In a preferred embodiment, the mono-unsaturated short chain fatty acid is preferably crotonic acid. Thus, the copolymer is preferably a copolymer of vinyl acetate and crotonic acid. A suitable copolymer is commercially available under the tradename WACKER VINNAPAS C305.

[030] The vinyl acetate / fatty acid copolymer functions within the hot melt adhesive to improve cohesive and adhesive strength for the formulation, especially when the adhesive is exposed to high humidity conditions.

[031] In some instance, the polymeric composition may also include one or more further additives. For instance, the polymeric composition may include one or more additives selected from the group consisting of fillers, pigments, anti-caking additives, defoaming agents, release additives, antioxidants, stabilizers, waxes, plasticizers, biocides, and anti-static additives.

[032] Preferably as many of the components of the hot melt adhesive as possible are derived from renewable resources. Such components may be referred to as being bio-sourced. It is also preferable that as many of the components of the hot melt adhesive as possible are biodegradable and/or recyclable. However, it is difficult to prepare a suitable hot melt adhesive using only these materials. Nonetheless, according to certain embodiments of the present disclosure, at least 50 weight percent of the components of the hot melt adhesive are bio-sourced. More preferably, at least 75 weight percent of the components of the hot melt adhesive are bio-sourced. In particular, at least the lactic acid oligomer or polymer and the polylactide are generally derived from renewable resources.

In some instances, the polyester may also be derived from renewable resources. For instance, PBSA may be derived from renewable resources.

[033] Further, according to certain embodiments of the present disclosure, at least 50 weight percent of the components of the hot melt adhesive are biodegradable. More preferably, at least 75 weight percent of the components of the hot melt adhesive are biodegradable. In particular, at least the lactic acid oligomer or polymer and the polylactide are generally biodegradable. In some instances, the polyester may also be a biodegradable polymer such as PBSA.

[034] In general, the hot melt adhesive composition is typically prepared by combining and melt blending its components. For instance, in a first (and in some instances single) step, the lactic acid oligomer or polymer, the polylactide, the diol / diacid polyester, and the copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid may be combined and melt blended together. The melt blending is generally carried out at a temperature sufficient to melt all of the components, typically from about 140 to about 175 °C.

[035] In general, it is believed that order of addition of the components when mixing does not affect the final properties of the adhesive composition. The method also includes a second step of further blending the mixture with from about 1 to about 5 weight percent of a carboxylic acid or a carboxylic acid derivative, at a temperature at a temperature from about 140 to about 175 °C to provide a stabilized hot melt adhesive. All of the aforementioned weight percentages are based on the overall weight of the hot melt adhesive.

[036] Optionally, in some instances, the method for preparing the hot melt adhesive may also include a second step of further blending the aforementioned mixture with from about 1 to about 5 weight percent of a carboxylic acid or a carboxylic acid derivative at a temperature at a temperature from about 140 to about 175 °C. Acid anhydrides are preferred carboxylic acid derivatives for this purpose, and propionic anhydride is particularly preferred. Addition of the carboxylic acid or a carboxylic acid derivative at these temperatures is believed to lead to reaction with the polylactide and/or the diol /

diacid polyester and to the formation of the aforementioned polymer capping group on the ends of the polylactide and/or the diol / diacid polyester molecules. The formation of these polymer capping groups is believed to improve the heat stability of the hot melt adhesive composition, as further described below.

[037] In some embodiments, either or both of the mixing steps may be carried out using a heated tank with a suitable agitator such as a high shear mixer for example. Alternatively, however, the components of the hot melt adhesive composition may be added to an extruder and heated and mixed within the extruder before being extruded through a die. If desired, the hot melt adhesive composition may be extruded directly onto a suitable substrate. More typically, however, the hot melt adhesive composition is initially extruded into pellets or any other desired form and then is allowed to cool and solidify. Once in a pelletized or other solid form, the hot melt adhesive may be packaged for storage and / or shipping. The pellets are eventually reheated and melted and applied to suitable substrate during a second extrusion step.

[038] Once prepared as described above, the hot melt adhesive of the present disclosure generally has a viscosity from about 1000 centipoise to about 6000 centipoise at a temperature of about 143°C. More preferably, the hot melt adhesive has a viscosity from about 2000 centipoise to about 4000 centipoise at a temperature of about 143°C, and most preferably a viscosity from about 2000 centipoise to about 3000 centipoise at a temperature of about 143°C.

[039] Advantageously, the hot melt adhesive composition has been found to exhibit improved stability when exposure to elevated temperatures. For instance, the hot melt adhesive composition has been found to remain viscosity stable for least 4 to 12 hours before significant degradation of the adhesive composition renders the composition unusable. Preferably, the hot melt adhesive composition retains a viscosity from about 4000 to about 5000 centipoise after being held at a temperature from about 135 °C to about 145 °C for a time period from about 8 to about 16 hours. More preferably, the hot melt adhesive composition retains a viscosity from about 4000 to about 5000 centipoise

after being held at a temperature from about 135 °C to about 145 °C for a time period of at least 48 hours.

[040] Once prepared, the hot melt adhesive composition is heated to a temperature of at least 140 °C in order to insure good flowability. The hot melt adhesive composition may then be applied to any desired substrate surface. For instance, the hot melt adhesive may be applied to paper or paperboard. Substrates to which the hot melt adhesive is applied may be used for both hot and cold food service items (such as plates, cups, and bowls) paperboard packaging, and carton or case seals including those used for frozen foods.

[041] Suitable methods for application of the hot melt adhesive to the substrate include extrusion nozzle application, hand gun application, roll coating application, and profile wrapping application.

[042] Once the hot melt adhesive is applied to the substrate and allowed to cool, the adhesive preferably exhibits good initial bond strength.

[043] EXAMPLES

[044] The following nonlimiting examples illustrate various additional aspects of the invention. Unless otherwise indicated, temperatures are in degrees Celsius and percentages are by weight.

[045] Example 1 & 2 – Preparation of Hot Melt Adhesive Compositions with End Capping.

[046] Example 1 – In this example, a hot melt adhesive composition was prepared. A two liter reactor equipped with a condenser was charged with the following components:

<u>Component</u>	<u>Weight (grams)</u>	<u>Weight percentage</u>
Poly lactide	1017 g	56.5%
Lactic acid oligomer	261 g	14.5%
Polybutylene (succinate-co-	468 g	26.0%

adipate) (“PBSA”)		
Wacker Vinnapas C305	54 g	3.0%
Total	1800 g	100.0%

[047] The polylactide had a weight average molecular weight of about 14,879. The lactic acid oligomer had a weight average molecular weight of about 1645. Vinnapas C305 is a copolymer of vinyl acetate and crotonic acid.

[048] The reactor pressure was reduced to 1 torr and allowed to sit overnight to remove any surface moisture. Under nitrogen, the reactor was then heated to 150 °C for 3 hours until all materials were melted and homogenously blended. Propionic anhydride (55.9 grams) was added to the reactor and allowed to stir for 2 hours, after which the pressure was slowly reduced down to 2-5 torr to remove any excess propionic anhydride. The adhesive product was collected as a tan solid. The weight average molecular weight (Mw) of the final adhesive was measured to be 36447 and the polydispersity was measured to be 4.02.

[049] Example 2 – A second hot melt adhesive was prepared. As in Example 1, a two liter reactor equipped with a condenser was charged with the following components:

<u>Component</u>	<u>Weight (grams)</u>	<u>Weight percentage</u>
Polylactide	904	56.5
Lactic acid oligomer	224	14.0
Polybutylene (succinate-co-adipate) (“PBSA”)	416	26.0
Wacker Vinnapas C305	56	3.5
Total	1600	100.0%

[050] The polylactide had a weight average molecular weight of about 18,323. The lactic acid oligomer had a weight average molecular weight of about 2609.

[051] Again, the reactor pressure was reduced to 1 torr and allowed to sit overnight to remove any surface moisture. Under nitrogen, the reactor was then heated to 150 °C for 3 hours until all materials were melted and homogenously blended. Succinic anhydride (51.9 grams) was added to the reactor and allowed to stir for 2 hours, after which the pressure was slowly reduced down to 2-5 torr to remove any excess propionic anhydride. The hot melt adhesive product was collected as a tan solid.

[052] Example 3 – Viscosity Testing.

[053] The hot melt adhesive of Example 1 was submitted to viscosity testing using a DV-II+ Brookfield viscometer equipped with a Thermosel for more accurate temperature control and a #27 aluminum spindle. In the testing procedure, 16 grams of the hot melt adhesive resin was added to an aluminum cup and placed in the Thermosel at 290 °F/ 143 °C. After 10 minutes of equilibration at 10 RPM, the viscosity of the material was recorded. Using this procedure, the initial viscosity of the adhesive (prior to aging at elevated temperature) was measured to be 5100 centipoise.

[054] The hot melt adhesive was then aged by being held at a temperature of 290 °F/143 °C for a total of 72 hours, with the viscosity being measured and recorded again at 24 hours, 48 hours, and 72 hours. The viscosity measurements are summarized in the following table:

[055] Adhesive Viscosity, Initial & After Aging

Time at 143°C (hours)	0	24	48	72
Viscosity (cP)	5100	5300	4125	2638

[056] These viscosity numbers demonstrate that the inventive hot melt adhesive exhibit both good initial viscosity and that this viscosity is substantially maintained when the hot melt adhesive composition is held at an elevated temperature for an extended period of time. This indicates that the components of the composition are slow to degrade under these elevated temperature conditions.

[057] Examples 4 – 7: Adhesive Testing.

[058] The adhesive properties of the hot melt adhesive of Example 1 were analyzed in a series of tests. The set time, open time, and the hot tack seal strength were each separately measured for the hot melt adhesive of Example 1. Each of the test were carried out using Hot Melt Tester, Model ASM-15N. Application temperature of the adhesive during the testing was 290 °F/ 143 °C.

[059] Example 4 – Set Time Testing

[060] The set time for the hot melt adhesive of Example 1 was first measured. As used herein, “set time” refers to the amount of time required for a bond to form after an adhesive is applied to a first substrate and a second substrate is pressed against it. For example, glue may be applied to a piece of cardboard and another piece of cardboard may then be applied over the adhesive and held for a few seconds. The set time is the minimum amount of time the two pieces must be held together to insure that the two piece are in fact bonded together and will not separate after the hold is released.

[061] The initial set time for the adhesive was measured, and then the set time was measured again after the hot melt adhesive was aged by being held at a temperature of 290 °F/ 143 °C for a total of 72 hours, with the set time being measured and recorded again at 24 hours, 48 hours, and 72 hours. The set time measurements are summarized in the following table:

[062] Adhesive Set Time, Initial & After Aging

Time at 143°C (hours)	0	24	48	72
Set time (second)	3.5 s	4 s	5 s	4.5 s

[063] These viscosity numbers demonstrate that the inventive hot melt adhesive exhibit both a favorable initial set time and that this set time does not degrade substantially when the hot melt adhesive composition is held at an elevated temperature for an extended period of time.

[064] Example 5 – Additional Set Time Testing

[065] Two additional samples of the hot melt adhesive of Example 1 were submitted for set time testing. Prior to measurement of the set time, the first sample was heated to 143°C, then removed from heat and allowed to cool to room temperature, and then reheated back to 143°C. The second sample was allowed to age overnight at 143°C in a humid environment of approximately 80% relative humidity prior to the set time measurement. The set time measurements are summarized in the following table:

[066] Adhesive Set Times – Heat/Cool/Reheat and High Humidity

Sample Conditions	Heated/Cooled/Reheated	Overnight at 80% RH
Set time (second)	3 s	4.5 s

[067] Example 6 – Open Time Testing

[068] The open time for the hot melt adhesive of Example 1 was also measured. As used herein, “open time” refers to the length of time after an adhesive is applied to a substrate in which a bond may be formed. For example, glue may be applied to a first piece of cardboard and after 5 seconds, another piece of cardboard may still be applied and still bonded to the first cardboard piece. But after 6 seconds, the glue may be too hard and set to form a bond between the two cardboard pieces. In this instance, the open time would then be 5 seconds.

[069] The open time for the adhesive was measured after the adhesive was heated to a temperature of 290 °F/ 143 °C. The open time was found to be 10 seconds.

[070] Example 7 – Hot Tack Testing

[071] The hot tack seal strength for the hot melt adhesive of Example 1 was also measured. As used herein, “hot tack seal strength” refers to the strength of heat seals formed between thermoplastic surfaces of flexible webs, immediately after the seal has been made and before it cools to room temperature. This measurement is of significance in high speed form-fill-seal packaging operations. Hot tack seal strength was measured

in accordance with ASTM F-1921. The hot tack was measured at both 0.5 second after seal formation and at 1.0 seconds after seal formation. The results are reported below:

Hot Tack Strength at 0.5 second	4.318 kg
Hot Tack Strength at 1.0 second	4.732 kg

[072] The hot tack seal strength numbers indicate that the inventive hot melt composition may suitably in high speed form-fill-seal packaging operations.

[073] Comparative Example 8 – Mechanical Testing of Technomelt 8370

[074] In this Example, samples of a conventional EVA hot melt adhesive (Technomelt 8370, available from Henkel) were subjected to adhesive testing in a manner similar to that described above in Examples 4 – 6.

[075] Multiple replicates were tested at three different temperatures (325 °F, 350 °F, and 375 °F) and averaged to determine the average set time and open time for the Technmelt adhesive. The results are summarized in the following table:

[076] Set Time and Open Time for Technmelt 8370

<u>Test No.</u>	<u>Hot Melt Application Temp</u>	<u>Ave. Set Time (s)</u>	<u>Ave. Open Time (s)</u>
1	176.4 °C (350 °F)	2.5 s	12 s
2	163 °C (325 °F)	2 s	9 s
3	190.5 °C (375 °F)	2.5 s	11 s

[077] As noted above, typical set times for the biodegradable hot melt of the present invention range from about 3 – 5 seconds, and typical open times for the biodegradable hot melt of the present invention to be about 10 seconds. Thus, it may be seen that the set times and open times for the inventive biodegradable are comparable to those for the conventional (non-biodegradable) Technomelt hot melt.

[078] The foregoing description of preferred embodiments for this invention has been presented for purposes of illustration and description. It is not intended to be exhaustive or to limit the invention to the precise form disclosed. Obvious modifications or variations are possible in light of the above teachings. The embodiments are chosen and described in an effort to provide the best illustrations of the principles of the invention and its practical application, and to thereby enable one of ordinary skill in the art to utilize the invention in various embodiments and with various modifications as are suited to the particular use contemplated. All such modifications and variations are within the scope of the invention as determined by the appended claims when interpreted in accordance with the breadth to which they are fairly, legally, and equitably entitled.

WHAT IS CLAIMED IS:

Claim 1. A hot melt adhesive comprising:

from 10 to 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from 1500 to 3000;

from 40 to 75 weight percent of polylactide having a weight average molecular weight from 10,000 to 18,000;

from 20 to 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids; and

from 0.5 to 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms,

wherein all weight percentages are based on the overall weight of the hot melt adhesive.

Claim 2. The hot melt adhesive of Claim 1, wherein the hot melt adhesive has a viscosity from 1000 centipoise to 6000 centipoise at a temperature of 143°C.

Claim 3. The hot melt adhesive of Claim 1, wherein the lactic acid oligomer or polymer has a viscosity from 350 centipoise to 450 centipoise at a temperature of 280°C.

Claim 4. The hot melt adhesive of Claim 1, wherein the polylactide has a viscosity from 1800 centipoise to 2200 centipoise at a temperature of 143°C.

Claim 5. The hot melt adhesive of Claim 1, wherein the polylactide further comprises a polymer capping group formed by reaction of the polylactide with a carboxylic acid or a carboxylic acid derivative.

Claim 6. The hot melt adhesive of Claim 1, wherein the polyester has a viscosity from 15,000 centipoise to 35,000 centipoise at a temperature of 216°C.

Claim 7. The hot melt adhesive of Claim 1, wherein the polyester has a weight average molecular weight from 55,000 to 72,000.

Claim 8. The hot melt adhesive of Claim 1, wherein the polyester further comprises a polymer capping group formed by reaction of the polylactide with a carboxylic acid or a carboxylic acid derivative.

Claim 9. The hot melt adhesive of Claim 1, wherein the polyester is polybutylene(succinate-co-adipate) ("PBSA").

Claim 10. The hot melt adhesive of Claim 1, wherein the mono-unsaturated short chain fatty acid is crotonic acid.

Claim 11. An adhesive-coated substrate comprising:

a substrate having at least a first side; and

a hot melt adhesive coating applied to at least a portion of the substrate first side, wherein the hot melt adhesive coating comprises

from 10 to 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from 1500 to 3000,

from 40 to 75 weight percent of polylactide having a weight average molecular weight from 10,000 to 18,000,

from 20 to 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids, and

from 0.5 to 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms,

wherein all weight percentages are based on the overall weight of the hot melt adhesive.

Claim 12. The adhesive-coated substrate of Claim 11, wherein the hot melt adhesive has a viscosity from 1000 centipoise to 6000 centipoise at a temperature of 143°C.

Claim 13. The adhesive-coated substrate of Claim 11, wherein the lactic acid oligomer or polymer has a viscosity from 350 centipoise to 450 centipoise at a temperature of 280°C.

Claim 14. The adhesive-coated substrate of Claim 11, wherein the polylactide has a viscosity from 1800 centipoise to 2200 centipoise at a temperature of 143°C.

Claim 15. The adhesive-coated substrate of Claim 11, wherein the polylactide further comprises a polymer capping group formed by reaction of the polylactide with a carboxylic acid or a carboxylic acid derivative.

Claim 16. The adhesive-coated substrate of Claim 11, wherein the polyester has a viscosity from 15,000 centipoise to 35,000 centipoise at a temperature of 216°C.

Claim 17. The adhesive-coated substrate of Claim 11, wherein the polyester has a weight average molecular weight from 55,000 to 72,000.

Claim 18. The adhesive-coated substrate of Claim 11, wherein the polyester further comprises a polymer capping group formed by reaction of the polylactide with a carboxylic acid or a carboxylic acid derivative.

Claim 19. The adhesive-coated substrate of Claim 11, wherein the polyester is polybutylene(succinate-co-adipate) ("PBSA").

Claim 20. The adhesive-coated substrate of Claim 11, wherein the mono-unsaturated short chain fatty acid is crotonic acid.

Claim 21. A method for making a hot melt adhesive, comprising the steps of:

melt blending, at a temperature from 140 to 175 °C, a mixture which comprises

from 10 to 20 weight percent of a lactic acid oligomer or polymer having a weight average molecular weight from 1500 to 3000,

from 40 to 75 weight percent of polylactide having a weight average molecular weight from 10,000 to 18,000,

from 20 to 35 weight percent of a polyester formed from the copolymerization of one or more diols and one or more dicarboxylic acids, and

from 0.5 to 5 weight percent of a copolymer of vinyl acetate and a mono-unsaturated short chain fatty acid, the fatty acid having from 4 to 12 carbon atoms; and

further blending the mixture with from 1 to 5 weight percent of a carboxylic acid or a carboxylic acid derivative, at a temperature at a temperature from 140 to 175 °C to provide a stabilized hot melt adhesive,

wherein all weight percentages are based on the overall weight of the hot melt adhesive.

Claim 22. The method of Claim 21, wherein the carboxylic acid or a carboxylic acid derivative comprises an acid anhydride.

Claim 23. The method of Claim 21, wherein the polyester has a weight average molecular weight from 55,000 to 72,000.

Claim 24. The method of Claim 21, wherein the polyester is polybutylene(succinate-co-adipate) ("PBSA").

Claim 25. The method of Claim 21, wherein the mono-unsaturated short chain fatty acid is crotonic acid.