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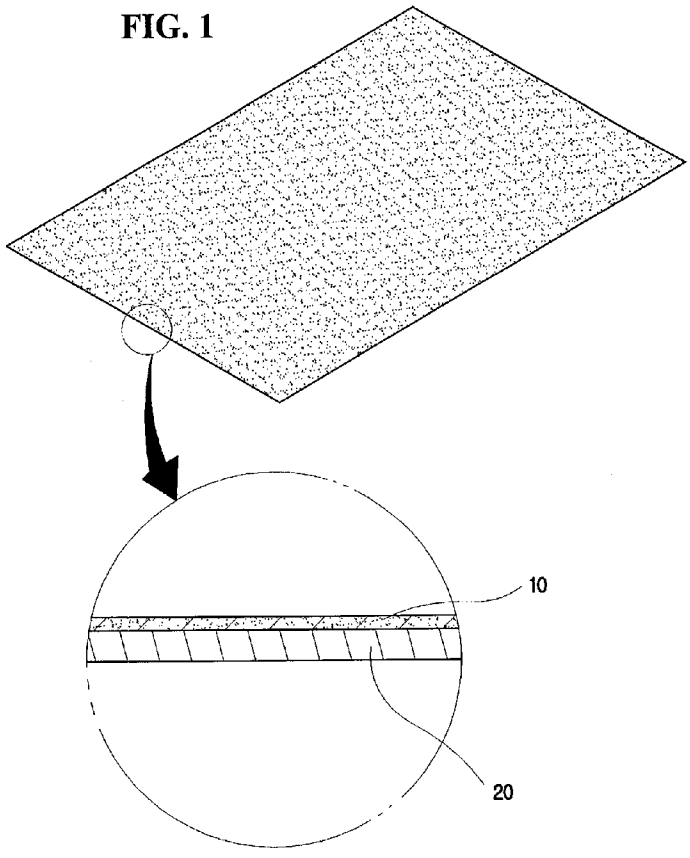
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(54) Title: THE COMPOSITION FOR LAMINATE COATING OF PAPER, AND THE FOOD PACKAGING PAPER USING IT

FIG. 1



(57) Abstract: Disclosed is a biodegradable resin composition for laminate coating of papers such as food packaging paper or a container. More particularly, disclosed are a biodegradable resin composition for laminate coating in which a previous problem that an environmental hormone detrimental to the human body was exited at a site for laminating a food packaging paper can be overcome and the food packaging paper can be biodegraded at a natural environment after its use by incorporating a highly degradable biodegradable polylactic acid (PLA) and a biodegradable polyester resin so that coating may achieved without an adhesive; and a food packaging paper employing the same.

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【DESCRIPTION】

【Invention Title】

THE COMPOSITION FOR LAMINATE COATING OF PAPER, AND THE FOOD PACKAGING PAPER USING IT

【Technical Field】

The present invention relates to a biodegradable resin composition for laminate coating of papers such as a food packaging paper or a container. More particularly, the present invention relates to a biodegradable resin composition for laminate coating which a previous problem that an environmental hormone detrimental to the human body was exited at a site for laminating a food packaging paper can be overcome and the food packaging paper can be biodegraded at a natural environment after its use by incorporating a highly degradable biodegradable polylactic acid (PLA) and a biodegradable polyester resin (= aliphatic polyester resin) so that coating may be achieved without an adhesive; and a food packaging paper employing the same.

【Background Art】

For conventional laminate coating paper, various synthetic resins such as PE (LDPE, HDPE), which is a general-purpose resin, and PP, PVC and URETHANE resin have been used. Harmful material such as a plasticizer or an adhesive is used in those synthetic resins to impart

plasticity or thermal adhesiveness thereto. It is known that such adhesive or some plasticizers elute a harmful environmental hormone greatly in hot water.

Most of laminate coating papers are now used in articles, which contact with food directly, such as a general cup, a noodle cup and a food tray that are disposable food products. In such circumstances, the elution of an environmental hormone reaches a serious level since particularly a cup noodle container or a paper cup is under the condition in which it contacts with hot water.

Further, a common resin may damage to the nature system since it is not degraded for decades due to its persistent properties when it is embedded. A persistent resin film may cause trouble in a process for recycling. Further, a PVC resin generates dioxin, and thus it is difficult to incinerate it.

Continuous research and application for PHA, PCL, PLA (polylactic acid), PHBV and PBS as a biodegradable resin for laminating on a paper have been achieved.

As a coating method employing a biodegradable resin up to date, there are disclosed a method of coating after dissolving a polyhydroxyalkanoate (PHA) resin in a solvent to form a solution (Korean Patent Application No. 1994-0011966), a method of coating by dispersing an aliphatic

polyester in water to form a solution (Korean Patent Application No. 2003-0041254), a method of coating by dissolving an aliphatic/aromatic copolyester resin in water to form a solution (Korean Patent Application No. 2005-0110211). The disclosed methods described above coat by forming a solution of a biodegradable resin. However, that resin has excellent biodegradability, but has shortcomings in that equipment for drying and recovering the solvent is needed, and its property of matter is weak.

Further, a melt coating method employing a PLA resin (Korean Patent Application No. 1999-7010885) and a melt coating method employing a biodegradable aliphatic polyester (Korean Patent Application No. 2000-0080964), etc. are disclosed. However, PLA has low expansibility, is very hard and has low thermal stability thereby causing fear that it pyrolyzed. The aliphatic polyester has also low thermal stability, is difficult to form a thin membrane thereby thickening its thickness, and is too soft to process.

The present inventors disclosed a method of forming a film by mixing a PLA resin and an aliphatic polyester (Korean Patent Application No. 2001-0073862). In the invention, it is possible to form a film alone, but it is difficult to process the film due to its low thermal stability when coated on a paper, and the film sticks to a

cooling roll due to low cooling speed, or is difficult to obtain a uniform coating surface.

In fact, PLA (polylactic acid) is in itself used as material for a general-purpose product through injection, extrusion or forming, etc., but it is known that it is difficult to use as material for laminating on a paper substrate due to its slow solidification speed and weak properties of matter despite the excellent properties. In particular, PLA has advantages that not only it has high degradability, but also it is not harmful to the human body, and its caloric value is low thereby generating dioxin or carbon dioxide in little amounts when burnt. However, a separate adhesive or double coating is needed on coating on a paper since PLA has weak adhesive force with a paper. Accordingly, costs rise, processes become complex and an environmental hormone due to use of an adhesive must be endured. As described below, the present invention overcame the above shortcomings by mixing other material together.

For coating a resin on a paper, there are a way of dissolving a resin with T-Die to form a film and simultaneously hot melt coating on a paper (corresponding to the laminate coating according to the present invention), a way of preparing a film for coating on a paper, and then adhering the film with an adhesive, and a way of dissolving

a resin to be coated in a solvent and applying it to a paper to coat, etc. PLA made it difficult to apply a hot melt manner in terms of its property, but the present invention achieved this.

【Disclosure】

【Technical Problem】

The object of the present invention is to provide a biodegradable resin composition for laminate coating of papers, which overcomes the previous problem that an environmental hormone harmful to the human body has been exited at a site for laminating a food packaging paper, as well as renders biodegradable in a natural environment by incorporating highly degradable polylactic acid and a biodegradable polyester resin (= aliphatic polyester resin), etc. so that coating may be achieved without an adhesive.

【Technical Solution】

A biodegradable resin composition for laminate coating of papers according to the present invention comprises 10 to 85% by weight of polylactic acid, 10 to 75% by weight of a biodegradable polyester resin (aliphatic polyester resin) and 0.05 to 5% by weight of a plasticizer. The composition may further comprise 0.01 to 50% by weight of an additive in which at least one or more of a stabilizer, a slipping agent, a dispersant, a coupling agent, an anti-oxidizing

agent, a UV stabilizer and a filler are mixed, relative to 100% by weight of the composition.

The polylactic acid is any one selected from the group consisting of a homopolymer of D-lactic acid, a homopolymer of L-lactic acid, or a copolymer of L-lactic acid and D-lactic acid; the plasticizer is at least one selected from the group consisting of sorbitan monolaurate, sorbitan monopalmitate, sorbitan trioleate, sorbitan monostearate, polyethylene glycol, polyalkylene oxide, glycerol, glycerol acetate, propylene glycol and sorbitol.

The biodegradable polyester resin may be any aliphatic polyester resin, and includes polybutylene adipate/terephthalate (trade name Ecoflex) or polybutylene succinate · co · adipate (trade name Enpol).

The filler may optionally employ at least one selected from the group consisting of talc, calcium carbonate, Limestone, TiO_2 and carbon black; the stabilizer may optionally employs at least one selected from the group consisting of trimethyl phosphate, phosphoric acid and triphenyl phosphate; the anti-oxidizing agent may optionally employ at least one selected from the group consisting of Irganox series, Ultranox series and TEP series; the slipping agent may optionally employ at least one selected from the

group consisting of calcium stearate, zinc stearate, PE wax and general wax.

The coupling agent may be anhydrous maleic acid and silane coupling agent (trimethoxysilane, triethoxysilane, tetraethoxysilane) etc.

The above resin composition is coated on a surface of papers having a thickness of 0.01 to 2 ㎜, thereby preventing moistures from entering and enhancing the exiting of moistures.

As described previously, PLA had a shortcoming in that its adhesive force with a paper is weak despite its advantages; however, the present invention overcomes the problem by mixing a biodegradable polyester resin (= aliphatic polyester resin) together. That is, the present invention removes the need of an adhesive harmful to the human body by incorporating a biodegradable polyester resin in order to enhance the adhesive force of PLA to a paper. Further, the present invention further incorporates a proper stabilizer considering that the biodegradable resin is broken when heated. The present invention also incorporates a proper coupling agent to overcome the problem that PLA and other biodegradable resin are immiscible and thus coating may not be properly achieved when the resin is used as such.

【Advantageous Effects】

When the biodegradable resin composition according to the present invention is used in a disposable food container (a paper cup, a cup noodle container, a disposable tray, etc.) as a biodegradable coating paper manufactured by coating the composition on a surface or both surfaces of papers under a predetermined condition, it can be widely applied as an eco-friendly coating paper that is not harmful to the human body since there is no an environmental hormone as well as does not cause an environmental pollution.

【Description of Drawings】

FIG. 1 illustrates the food packaging paper according to the present invention.

【Mode for Invention】

The present invention provides a biodegradable resin composition for laminating on various food packaging papers comprising 10 to 85% by weight of polylactic acid, 10 to 75% by weight of a biodegradable polyester resin (aliphatic polyester resin) and 0.05 to 5% by weight of a plasticizer. Herein, a polylactic acid, a polyester resin, a plasticizer and a filler are further incorporated. The polylactic acid (PLA) is a biodegradable resin different from a general-purpose synthetic resin product, and is not harmful to the human body and does not change the physical strength of a paper. Further, the polylactic acid has a characteristic

that can overcome a problem that previous PE or PP is not degraded thereby remaining wastes in the earth due to its biodegradability at natural state. The present invention uses any one selected from the group consisting of a homopolymer of D-lactic acid, a homopolymer of L-lactic acid or a copolymer of L-lactic acid and D-lactic acid as the polylactic acid.

In the present invention, a biodegradable polyester resin is further incorporated. The resin is obtained by performing polycondensation reaction after undergoing any one or more than two reaction selected from (1) polycondensation, (2) ester exchange reaction, or (3) esterification reaction. When the resin is incorporated into the composition, it prevents moisture from entering and helps exiting vapors. The composition according to the present invention is laminated on the surfaces of various food packaging papers or containers. For a noodle container on which the composition is laminated, the composition plays a role so that papers do not absorb moistures even when hot water is poured although materials are papers. Accordingly, even though the container is manufactured with a paper, it can contain water.

In the present invention, a plasticizer is further incorporated. The plasticizer generally refers to an

organic material that facilitates forming process at high temperatures by enhancing thermoplastic property when added to a thermoplastic material. It is the characteristic of the plasticizer to enhance heat resistance, cold resistance, flame resistance and electrical property, etc. in addition to compatibility and plasticity. The plasticizer used most is DOP (dioctyl phthalate), DOA (dioctyl adipate) and TCP (tricresyl phosphate). However, the present invention optionally mixes and uses at least one or more selected from the group consisting of sorbitan monolaurate, sorbitan monopalmitate, sorbitan trioleate, sorbitan monostearate, polyethylene glycol, polyalkylene oxide, glycerol, glycerol acetate, propylene glycol and sorbitol as a plasticizer. These materials are also used as food additives, and are not harmful to the human body.

Finally, in the present invention, a separate filler can be incorporated to the mixture. The filler may optionally employ at least one selected from the group consisting of talc, calcium carbonate, Limestone, TiO_2 and carbon black, and is added in order to increase blocking phenomenon when processed and cooling speed. The average particle diameter of the filler is controlled in the range of 0.01 to 10 ㎛, and the filler can be used in an amount of

0.01 to 50% by weight when the above resin composition is referred to 100% by weight.

The resin composition prepared as above is applied on a food packaging paper or the surface of a packaging container, and hardened thereby forming a coated layer.

Meanwhile, the composition according to the present invention can further comprise the following additives. That is, the composition may further comprise 1 to 20% by weight of an additive in which at least one or more of a stabilizer, a slipping agent, a dispersant, a coupling agent, an anti-oxidizing agent and a UV stabilizer are optionally mixed, relative to 100% by weight of the composition. The stabilizer may optionally employ at least one selected from the group consisting of trimethyl phosphate, phosphoric acid and triphenyl phosphate in an amount of 0.01 to 10% by weight, relative to 100% by weight of the resin composition.

Further, Irganox series, Ultranox series and TEP series can be used as the anti-oxidizing agent in an amount of 0.01 to 10% by weight, relative to 100% by weight of the resin composition, and HALS (hindered amine light stabilizer) series can be used as the UV stabilizer in an amount of 0.01 to 10% by weight, relative to 100% by weight of the resin composition; calcium stearate, zinc stearate,

PE wax and general wax can be used as the slipping agent in an amount of 0.01 to 10% by weight, relative to 100% by weight of the resin composition.

In addition, carboxylated polyethylene, phthalic acid and stearic acid, etc. can be used as the dispersant that is an additive to be added for compatibility between the used resins. The dispersant can be used in an amount of 0.01 to 10% by weight, relative to 100% by weight of the resin composition.

As described above, the additives used in the present invention can be optionally added depending on the need of a product such as dispersibility or thermal stability.

In the present invention, an extruder (T-die) used in coating a biodegradable resin may be preferably downward type including T-shaped Die. There was a problem that much load of resin pressure are generated, and the extruded amount becomes less thereby decreasing product efficiency in the T-Die used in coating a previous synthetic resin. In order to improve such problem, the present invention enlarged the diameter of the previous orifice tube to 5 to 10 ㎜ to lessen the load of resin pressure, and improved the discharged amount to improve the same extruded amount of resin and productivity as the previous synthetic resin. Further, the temperature distribution of the screw and T-Die

was set at 150 to 350℃ considering the pyrolysis temperature of a biodegradable PLA and aliphatic polyester, and the problem of the previous paper coating of a biodegradable resin was overcome to improve the adhesive force with a paper substrate, coating speed and uniform coating thickness.

Various food-packaging papers are manufactured employing the resin composition prepared as described above. This is achieved by coating the resin composition in liquid state on the surfaces of various papers and simultaneously hardening.

The food packaging paper according to the present invention includes a coating layer 10 constituting upper layer by a composition for laminating, and papers 20 having a thickness of 0.01 to 2 ㎜. Accordingly, the coating layer is applied on the surface of the papers thereby preventing moistures from entering and enhancing the exiting of moistures. This is a simple constitution that a resin composition is coated on various kinds of papers. Herein, the thickness is preferably limited to 0.01 to 2 ㎜ since the durability of the coating layer becomes weak in proportion to the thickness of papers if it is too thin or too thick.

Preparation example

The amount of all components including additives (a slipping agent, an anti-oxidizing agent and a filler) is based on 100% by weight of the resin composition in the preparation example below.

1) 1.5% by weight of calcium stearate as a slipping agent and 1.5% by weight of n-octadecyl-β-(4'-hydroxy-3',5'-di-tert-butylphenyl) propionate(trade name Irganox 1076) as an anti-oxidizing agent were added to 97% by weight of polylactic acid [PLA, NatureWorks LLC, USA, Grade : 2002D, Melt Index 5~7], and the mixture was subjected to a twin screw extruder to prepare a composition in pellet.

2) 20% by weight of polybutylene adipate/terephthalate (BASF Corporation, trade name Ecoflex, grade : F BX 7011) as a biodegradable polyester resin, 0.5% by weight of polyethylene glycol as a plasticizer, 1.5% by weight of calcium stearate as a slipping agent, 1.5% by weight of n-octadecyl-β-(4'-hydroxy-3',5'-di-tert-butylphenyl) propionate (trade name Irganox 1076) as an anti-oxidizing agent were added to 76.5% by weight of polylactic acid [PLA, NatureWorks LLC, USA, Grade : 2002D, Melt Index 5~7], and the mixture was subjected to a twin screw extruder to prepare a composition in pellet.

3) 10% by weight of polybutylene succinate · co · adipate [trade name Enpol, Melt Index below 4, 2160g/10min] and 10% by weight of polybutylene adipate/terephthalate (BASF Corporation, Ecoflex, grade : F BX 7011) as biodegradable polyester resins, 0.3% by weight of polyethylene glycol as a plasticizer, 1.5% by weight of calcium stearate as a slipping agent and 1.5% by weight of n-octadecyl-β-(4'-hydroxy-3',5'-di-tert-butylphenyl) propionate (trade name Irganox 1076) as an anti-oxidizing agent were added to 76.7% by weight of polylactic acid [PLA, NatureWorks LLC, USA, Grade : 2002D, Melt Index 5~7], and the mixture was subjected to a twin screw extruder to prepare a composition in pellet.

4) 30% by weight of polybutylene succinate · co · adipate [trade name Enpol, Melt Index below 4, 2160g/10min] as a biodegradable polyester resin, 10% by weight of calcium carbonate (particle size 2.5 ㎛) as a filler, 0.2% by weight of polyethylene glycol as a plasticizer, 1.5% by weight of calcium stearate as a slipping agent and 0.5% by weight of n-octadecyl-β-(4'-hydroxy-3',5'-di-tert-butylphenyl) propionate (trade name Irganox 1076) as an anti-oxidizing agent were added to 57.8% by weight of polylactic acid [PLA, NatureWorks LLC, USA, Grade : 2002D, Melt Index 5~7], and

the mixture was subjected to a twin screw extruder to prepare a composition in pellet.

5) 40% by weight of polybutylene succinate·co·adipate [trade name Enpol, Melt Index below 4, 2160g/10min] and 10% by weight of polybutylene adipate/terephthalate (BASF Corporation, Ecoflex, grade : F BX 7011) as biodegradable polyester resins, 0.1% by weight of polyethylene glycol as a plasticizer, 1.5% by weight of calcium stearate as a slipping agent and 1.5% by weight of n-octadecyl-β-(4'-hydroxy-3',5'-di-tert-butylphenyl) propionate (trade name Irganox 1076) as an anti-oxidizing agent were added to 46.9% by weight of polylactic acid [PLA, NatureWorks LLC, USA, Grade : 2002D, Melt Index 5~7], and the mixture was subjected to a twin screw extruder to prepare a composition in pellet.

6) 1.5% by weight of n-octadecyl-β-(4'-hydroxy-3',5'-di-tert-butylphenyl) propionate (trade name Irganox 1076) as an anti-oxidizing agent were added to 67% by weight of polybutylene succinate·co·adipate [trade name Enpol, Melt Index below 4, 2160g/10min] and 20% by weight of polybutylene adipate/terephthalate (BASF Corporation, Ecoflex, grade : F BX 7011) as biodegradable polyester resins, 10% by weight of calcium carbonate (particle size 2.5 ㎛) as a filler and 1.5% by weight of calcium stearate as

a slipping agent, and the mixture was subjected to a twin screw extruder to prepare a composition in pellet.

7) 1.5% by weight of n-octadecyl-β-(4'-hydroxy-3',5'-di-tert-butylphenyl) propionate (trade name Irganox 1076) as an anti-oxidizing agent were added to 97% by weight of polybutylene succinate·co·adipate [trade name Enpol, Melt Index below 4, 2160g/10min] as a biodegradable polyester resin, and 1.5% by weight of calcium stearate as a slipping agent, and the mixture was subjected to a twin screw extruder to prepare a composition in pellet.

Example

The pellets prepared according to the above preparation example were heated, melted and mixed, and then melt-extruded using T-shaped Die and simultaneously laminated on papers. The resulting papers were evaluated for formability based on the following standard, and the results are shown in Table 1 below (Table 1 indicates main components only).

(Evaluation for formability)

○ : papers and coating are good, and the coated face is in excellent state since there was no thick or thin coated face

△ : papers and coating are good, and the coated face is in good state

× : papers and coating are bad, and the coated face is in bad state

Table 1

Item			Ex.1	Ex.2	Ex.3	Ex.4	Ex.5	Ex.6	Ex.7
Component		Unit							
Polylactic acid (PLA)		Wt%	97	76.5	76.7	57.8	46.9	0	0
Biodegradable polyester resin	Enpol	Wt%	0	0	10	30	40	70	97
	Ecoflex	Wt%	0	20	10	0	10	20	0
Filler ($CaCO_3$)		Wt%	0	0	0	10	0	10	0
Plasticizer		Wt%	0	0.5	0.3	0.2	0.1	0	0
Formability			×	△	△	○	○	△	×
Rheology right after extrusion				Very high	High	Mid	Mid	Soft	Very soft
Hardness after cooling			Not formed	High	Mid	Mid	Soft	Soft	Very soft

* Evaluation for coating characteristic and formability characteristic according to the mixing ratio of polylactic acid and a biodegradable polyester resin.

* When polylactic acid is used alone (Example 1), adhesion to a paper was difficult due to high cooling speed after extrusion, however, the property of matter satisfactory for post-processing was obtained by lowering cooling speed and the hardness of a film after extrusion through adding a biodegradable polyester resin.

* If the amount of polylactic acid is too low (Examples 6 and 7), the formability was lowered due to excessively low cooling speed, while if the amount of

polylactic acid is too high (Examples 2 and 3), the adhesive force was weakened.

* The used plasticizer played a role in lowering thickness on extrusion by enhancing compatibility of polylactic acid and polyester resin, and was used properly in an amount of 0.05 to 5.0% by weight.

【Industrial Applicability】

A biodegradable resin composition for laminate coating according to the present invention, and a food packaging paper employing the same can be widely applied in the industry since they do not exit an environmental hormone harmful to the human body and is biodegradable in a natural environment.

Although the present invention has been described with reference to several embodiments of the invention, the description is illustrative of the invention and is not to be construed as limiting the invention. Various modifications and variations may occur to those skilled in the art, without departing from the spirit and scope of the invention as defined by the appended claims.

【CLAIMS】

【Claim 1】

A biodegradable resin composition for laminate coating of papers, the composition comprising 10 to 85% by weight of polylactic acid, 10 to 75% by weight of a biodegradable polyester resin (aliphatic polyester resin) and 0.05 to 5% by weight of a plasticizer.

【Claim 2】

The biodegradable resin composition of claim 1, wherein the composition further comprises 0.01 to 50% by weight of an additive in which at least one or more of a stabilizer, a slipping agent, a dispersant, a coupling agent, an anti-oxidizing agent, a UV stabilizer and a filler are mixed, relative to 100% by weight of the composition of claim 1.

【Claim 3】

The biodegradable resin composition of claim 1, wherein the polylactic acid is any one selected from the group consisting of a homopolymer of D-lactic acid, a homopolymer of L-lactic acid, or a copolymer of L-lactic acid and D-lactic acid.

【Claim 4】

The biodegradable resin composition of claim 1, wherein the plasticizer is at least one selected from the

group consisting of sorbitan monolaurate, sorbitan monopalmitate, sorbitan trioleate, sorbitan monostearate, polyethylene glycol, polyalkylene oxide, glycerol, glycerol acetate, propylene glycol and sorbitol.

【Claim 5】

The biodegradable resin composition of claim 2, wherein the filler is at least one selected from the group consisting of talc, calcium carbonate, Limestone, TiO_2 and carbon black.

【Claim 6】

The biodegradable resin composition of claim 2, wherein the stabilizer is at least one selected from the group consisting of trimethyl phosphate, phosphoric acid and triphenyl phosphate.

【Claim 7】

The biodegradable resin composition of claim 2, wherein the anti-oxidizing agent is at least one selected from the group consisting of Irganox series, Ultranox series and TEP series.

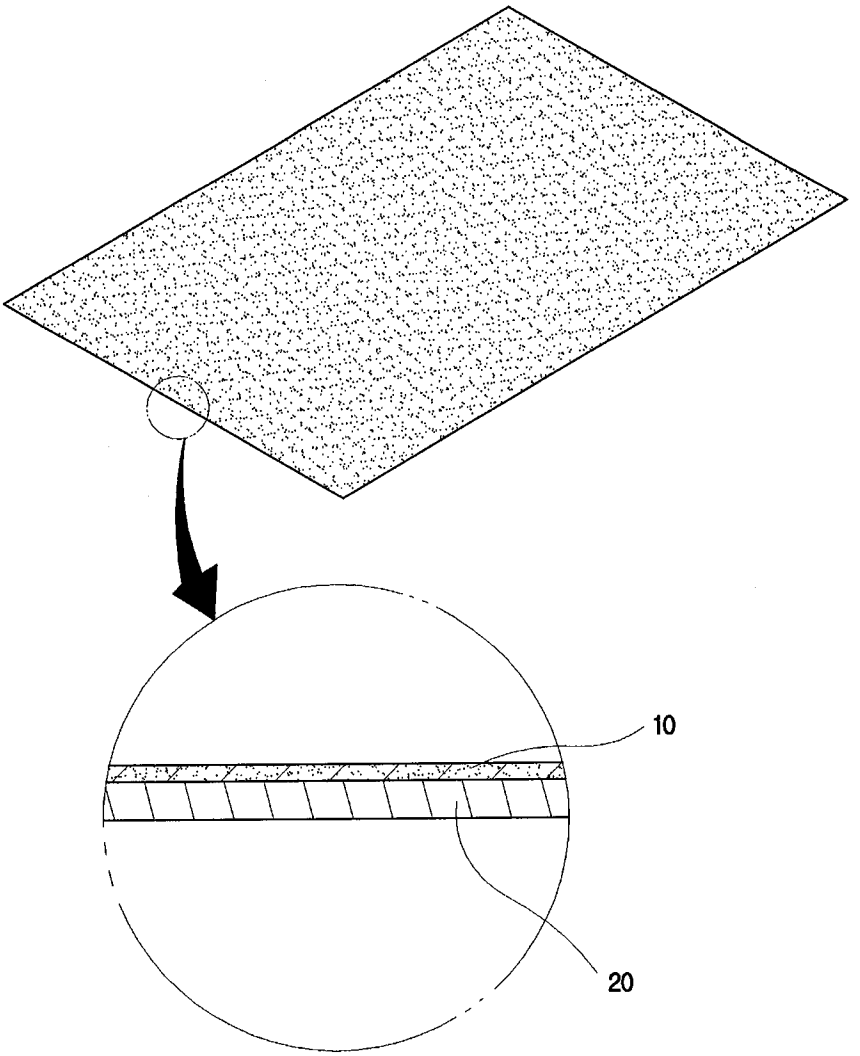
【Claim 8】

The biodegradable resin composition of claim 2, wherein the slipping agent is at least one selected from the group consisting of calcium stearate, zinc stearate, PE wax and general wax.

【Claim 9】

A food packaging paper on which a biodegradable resin composition for laminate coating of papers is applied, wherein a coating layer 10 comprised of the composition according to any one of claims 1 to 8 is formed on the surfaces of papers 20 having a thickness of 0.01 to 2 ㎜ thereby preventing moistures from entering and enhancing the exiting of moistures.

FIG. 1



INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2008/002981

A. CLASSIFICATION OF SUBJECT MATTER

C08L 67/04(2006.01)i

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 8 : C08L 67/04

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

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

eKIPASS; PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	KR 10-2003-0042945 A (SIM, JAE HO) 02 June 2003 See the abstract; page 2; claims 1-12.	1-9
Y	KR 10-2006-0003580 A (TORAY SAEHAN INC.) 11 January 2006 See the abstract; page 2 lines 33-36; page 3lines 1-2, 34-40; claims 1,2.	1-9
A	KR 10-2001-0066970 A (JOUNG, HYO SIK et al.) 12 July 2001 See the whole document.	1-9
A	US 5,753,724 A (Garry J. Edgington et al.) 19 May 1998 See the whole document.	1-9

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:

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INTERNATIONAL SEARCH REPORT

Information on patent family members

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