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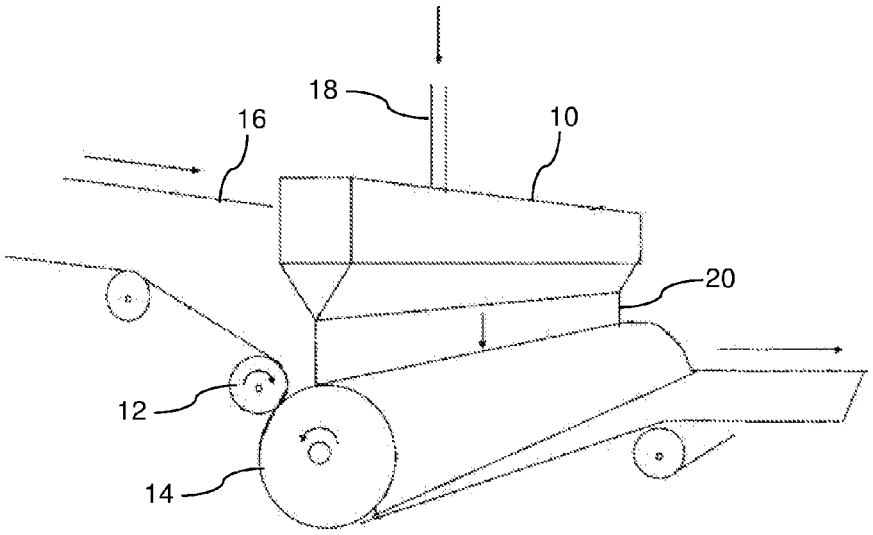


Fig. 1

(57) Abstract: A method of producing a laminate structure comprises the steps of receiving a web of high-density paper with a specially chemically and/or mechanically treated paper surface, wherein the density of the high-density paper is ≥0.8 grams per cubic centimetre, extrusion coating a layer of readily crystallisable polyolefin polymer resin or a multitude of layers of polyolefin resins onto the web of high-density paper, and applying pressure onto the polyolefin polymer resin coated high-density paper and in this way inducing the crystallization of the polymer, thereby forming a highly crystalline portion in the polymer layer, which polymer layer shows high gas and aroma barrier properties.

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A LAMINATE STRUCTURE

This invention relates to a method of producing a laminate structure and to the laminate structure itself.

It is very well-known in the packaging industry to package food and liquid food products (such as milk or fruit juice) in containers that are constructed from laminate material. Such laminate materials are usually created using a substrate such as paperboard or aluminium that is layered with one or more layers of polymer resin on one or both sides of the substrate. The choice of substrate and polymer layers will depend on the specific use of the laminate material and will take into account requirements such as desired stiffness/flexibility, oxygen barrier properties, moisture barrier properties and so on. Other factors that will be taken into account in the selection of the materials for the laminate structure include the cost of the materials used, the recyclability of the materials used and so on. There is a constant desire to produce a laminate material that is as cheap as possible, while providing the necessary barrier functions that compare with or exceed the properties of existing laminates.

It is therefore an object of the invention to improve upon the known art.

According to a first aspect of the present invention, there is provided a method of producing a laminate structure, the method comprising the steps of receiving a web of high-density paper formed by using refined chemical pulp and by subsequently treating the paper surface with alginic acid and/or starch and/or carboxyl methylcellulose and/or mechanical super calendaring and comprising a thickness of between 35 and 80 micrometre, a density of between 0.80 and 1.00 gr/cm^3 and a weight of between 30 and 80 gr/m^2, extrusion coating a layer of readily crystallisable polyolefin polymer resin onto the web of high-density paper, and applying a pressure of between 2 and 20

bar onto the layer of polyolefin polymer resin coated high-density paper thereby inducing crystallization in the portion of the polyolefin polymer resin adjacent to the high density paper, producing a level of crystallinity which is at least 15% higher than the remainder of the polyolefin polymer resin.

According to a second aspect of the present invention, there is provided a laminate structure comprising a substrate of high-density paper, formed by using refined chemical pulp and by subsequently treating the paper surface with alginic acid and/or starch and/or carboxyl methylcellulose and/or mechanical super calendaring and comprising a thickness of between 35 and 80 micrometre, a density of between 0.80 and 1.00 gr/cm^3 and a weight of between 30 and 80 gr/m^2, and a layer of readily crystallisable polyolefin polymer resin that has been extrusion coated onto the web of high-density paper, the polyolefin polymer resin layer including crystallization in the portion of the polyolefin polymer resin adjacent to the high density paper, with a level of crystallinity which is at least 15% higher than the remainder of the polyolefin polymer resin.

Owing to the invention, it is possible to provide, through the extrusion coating of a layer of readily crystallisable polyolefin polymer resin on high-density paper and the application of pressure, a multi-purpose packaging laminate structure showing good mechanical, aroma and gas barrier properties. The laminate structure is suitable for use as a packaging material usable on horizontal and vertical flow-wrapper machines, on pouch making machines including stand-up pouches machines, on thermoforming machines as top webs and on tray sealing equipment as lidding film.

Although high-density paper has basic properties like stiffness, oil-grease resistance, low air permeation rate and foldability that makes paper suitable for the use on automatic packaging machines, high-density paper lacks any notable form of water vapour, aroma or gas barrier and also has no sealing properties making high-density paper as such not having the necessary properties that are needed for use on automatic packaging machines. Polyolefin polymers on the other hand are extrusion coatable polymer resins that show properties such as water vapour barrier and

sealability while having generally insufficient barrier properties for gasses like O_2, N_2 and CO_2 and for volatile aromatic substances.

High-density paper can be characterized as paper made using extensively refined chemical pulp. The extensive refining of the fibres allows them to come more closely together in the paper structure and form more bonding sites and generally allow only the formation of smaller pores (<0.3 nanometre). In addition, the surface of the high-density paper is treated with alginic acid and/or starch and/or carboxyl methylcellulose and/or super calendared to work the high-density paper surface and give it a typical stereo metric surface configuration. The high-density paper is further characterized by a paper thickness range of between 35 micrometre and 80 micrometre, by a density of between 0.80 and 1.00 gr/cm^3 and by a grammage (m^2 weight) of between 30 and 80 gr/m^2.

The polyolefin polymer resins are all characterized by their readiness to crystallize and preferably have densities between 0.870 and 0.970 gr/cm^3 and can be applied in individual layers of between 4 and 80 gr/m^2 coating weight. The polyolefin polymer resin extrusion coated layer or layers can be HDPE (high density polyethylene), LDPE (low density polyethylene), MDPE (medium density polyethylene), LLDPE (linear low density polyethylene), LMDPE (linear medium density polyethylene), POP (polyolefin plastomer), EVA (ethylene vinyl acetate), EMA (ethylene methyl acrylate), EEA (ethylene ethyl acrylate), EBA (ethylene butyl acrylate), EAA (ethylene acrylic acid), or zinc-, sodium- or lithium based ionomer resins. Multi-layer combinations, or mixtures or blends of more than one of the above listed extrudable polymer resins can also be. Optionally, the listed resins can have inorganic fillers such as $CaCO_3$ or talcum added, for example.

In a preferred embodiment of an extrusion coating process, the polymer resin or multitude of polymer resins, is/are molten in one or more extruders and pumped as a melt stream into a film die where the film die converts the melt stream into a melt curtain that is extrusion coated on the high-density paper substrate. Subsequently the extrusion coated high-density paper with the polymer resin layer or layers will be wound on a roll in the wind up.

The polyolefin extrusion coated high-density paper structure has good machinability on (food) packaging machines such as flow wrappers (HFFS & VFFS), thermoforming machines, tray sealers and pouch making machines including stand-up machines. The laminate structure has sealability by heat, by ultrasonic, by induction and by high frequency. The structure has a water vapour barrier with WVTR (water vapour transmission rates) of typically < 5 gram H_2O per m^2 for 24 hours at 23°C and 85% RH (DIN 53122). The structure has a gas and aroma barrier with OTR rates (oxygen transmission rates) of typically < 10 cc O_2 per m^2 for 24 hours at 23°C and 50% RH (ASTM D 3985). The laminate structure also has good printability using one or more of different printing techniques commonly used in the trade (Flexo, Gravure, Offset, etc.).

The high-density paper alone has no gas or water vapour barrier. The polymer resins alone have no notable gas barrier but only a water vapour barrier. The extrusion-coated structure though that combines the polymer layer on the surface of the high-density paper has a good oxygen barrier. This is achieved without the use of dedicated barrier resins. The mechanism behind this is that in the interface between the high-density paper and the polyolefin a thin highly crystalline polymer layer is formed during the extrusion coating process using dedicated coating line parameters and which thin highly crystalline polymer layer gives the gas-barrier properties in combination with the paper itself that after coating does not have any pores anymore but only relative high gas-barrier cellulose fibres.

Embodiments of the present invention will now be described, by way of example only, with reference to the accompanying drawings, in which:-

Figure 1 is a schematic diagram of an extrusion coating line,

Figure 2 is a section through a laminate structure,

Figure 3 is a table showing different laminate structures,

Figure 4 is a flowchart of a method of producing the laminate structure, and

Figure 5 is a section through a laminate structure.

Figure 1 shows an extrusion coating line, of which the principal components are a film die 10, a nip roller 12 and a quench roller 14. A web 16 of high-density paper is received, which is the substrate for the laminate material that is produced by the extrusion coating line. A hot melt stream 18 is provided to the film die 10, which outputs a melt curtain 20 for depositing onto the web 16. The nip roller 12 is to provide pressure onto the surface of the web 16, after the polymer resin 20 has been deposited onto the web 16. The quench roller 14 is water-cooled and cools the web 16 after the resin 20 has been deposited.

In this way, a roll of high-density paper is unwound and fed through the nip roller 12 and quench roller 14 and a laminate structure is formed by the extrusion coating of the polymer resin 20 onto the web 16 of high-density paper. The output of the coated high-density paper is then wound back up into a roll for subsequent onward transport. The laminated high-density paper can then be converted into container blanks or used directly on a form-fill-seal machine, for example. This will depend upon the use to which the laminate structure is to be put, whether used for pouches or liquid food containers, etc.

The film die 10 shown in Figure 1 is designed to coat one single polymer resin 20 onto the web 16, but more complex dies 10 can be used. For example two different polymer resins can be coated onto the web 16 at the same time, from the same die 10. The internal construction of the die 10 is such that it contains different tanks that receive different melt streams 18 which are then guided into different melt curtains 20 that meet only at the exit f the die 10 and are coated onto the web 16 simultaneously. More complex dies 10 are also possible that will extrusion coat more than two different polymers at the same time.

The high-density paper used in the process is paper made using extensively refined chemical pulp. The extensive refining of the fibres allows the fibres to come more closely together in the paper structure and form more bonding sites and generally allow only the formation of smaller pores (<0.3 nanometre). In addition, the surface of the high-density paper is treated with

alginic acid and/or starch and/or carboxyl methylcellulose and/or super calendared to work the high-density paper surface. The high-density paper is further characterized by a paper thickness range of between 35 micrometre and 80 micrometre, by a density of between 0.80 and 1.00 gr/cm^3 and by a grammage (m^2 weight) of between 30 and 80 gr/m^2.

Figure 2 shows a section through the laminate structure 26 produced by the extrusion coating line of Figure 1. The laminate structure 26 comprises a substrate 22 of high-density paper, where the density of the high-density paper is greater than or equal to 0.8 grams per cubic centimetre (and preferably less than or equal to 1 gram per cubic centimetre) and a layer 24 of highly crystallisable polyolefin polymer resin, which has been extrusion coated onto the substrate 22. In a preferred embodiment, the polyolefin polymer resin 24 comprises a polyethylene polymer, an ethylene acetate polymer or an ethylene acrylate polymer or a mix or blend of such polymers. The polymer resin 24 could be HDPE (high density polyethylene) for example.

The high-density paper 22 has a thickness range of between 35 nanometres and 80 nanometres and the polymer resin 24 has a coating weight range of between 4 and 80 grams per square metre. The polymer resin 24 is extrusion coated onto the paper 22 and is then put under pressure by the nip roller 12 in order to bond the two substances together, in order to form the final laminate structure 26, shown in Figure 2. As discussed above, more complex laminate structures 26 can be created, using additional layers of high-density paper 22 and/or polymer resin 24, but the simplest form of the laminate structure 26 is shown in Figure 2.

The two materials together, the high-density paper 22 and the polymer resin 24, form a very effective packaging material. The laminate structure 26 is relatively straightforward to manufacture using readily available materials and has good stiffness and sealability properties (using heat or ultrasound etc.), while also providing good barrier properties in the important areas of water vapour, gas and aroma barriers. The laminate structure 26 is suitable for use in food packaging and can be used on thermoforming machines and tray sealing machines, for example. The high-density paper 22 can be printed on

before any extrusion coating takes place and is ideal for use in modern food and liquid food packaging systems.

Figure 3 shows a table of various different laminate structures 26, labelled L1 to L5. In this table of the structures "Polymer Resin" stands for one or more layers of extrusion coated polymer resins, for example taken from the list of twelve above mentioned polymer resins and "HD Paper" stands for one ply of high-density paper. This table shows that the polymer resin 24 can be extrusion coated on both sides of the high-density paper 22 and that multiple layers of the high-density paper 22 can be used in the laminate structure 26. The choice of which polymer resin(s) 24 and the number of layers of high-density paper 22 to be used will depend upon the ultimate application of the laminate structure 26.

Different polymer resins 24 can be used in different layers of the laminate structure 26 and again this will depend upon the ultimate purpose of the laminate structure 26 that is being created. Since multiple different layers can be created from the same die 10, as discussed above, then the actual layer referred to as "Polymer Resin" in the table of Figure 3 could actually be comprised of two or more individual layers of polymer resin 24 that have extrusion coated from the same die 10. Each "Polymer Resin" layer could also be comprised of a blend or mix of polymers that are essentially a single layer extrusion coated as such.

Inorganic fillers can also be used in the polymer resin layer(s) 24, again depending upon the purpose of the laminate structure 26 being created. $CaCO_3$ and talcum can be used, for example, as inorganic fillers in any of the layers of polymer resin 24. The film die 10 holds the molten resin(s) prior to the extrusion coating on the web 16, which are then coated onto the web 16 and immediately put under pressure by the nip roller 12, which presses the coated web 16 against the quench roller 14, which is being water cooled to harden the compressed resin(s) 24 in place. Multiple stations may be used in the extrusion coating line, in order to coat all of the necessary resin layers 24 in the laminate structure 26.

Figure 4 shows a flowchart that summarises the method of producing the laminate structure 26. The method comprises three principal steps, S1 to S3. Step S1 comprises receiving the web 16 of high-density paper 22, where the density of the high-density paper is greater than or equal to 0.8 grams per cubic centimetre. Step S2 comprises extrusion coating a layer of readily crystallisable polyolefin polymer resin 24 onto the web 16 of high-density paper 22, and step S3 comprises applying pressure of between 2 and 20 bar onto the polyolefin polymer resin coated high-density paper. These three steps take place in the extrusion coating line shown in Figure 1. The die 10 provides the resin 24 and the nip roller 12 provides the pressure.

The high-density paper is formed by using refined chemical pulp and by subsequently treating the paper surface with alginic acid and/or starch and/or carboxyl methylcellulose and/or mechanical super calendaring and has the following physical properties: a thickness of between 35 and 80 nm, a density of between 0.80 and 1.00 gr/cm^3 and by a weight of between 30 and 80 gr/m^2. The result of extrusion coating a layer of readily crystallisable polyolefin polymer resin onto the web of high-density paper and applying a pressure of between 2 and 20 bar onto the layer of polyolefin polymer resin coated high-density paper is induce crystallization in the portion of the polyolefin polymer resin adjacent to the high density paper, producing a level of crystallinity which is at least 15% higher than the remainder of the polyolefin polymer resin.

In the preferred embodiment, the step S3 of applying pressure onto the polyolefin polymer resin coated high-density paper occurs directly after the extrusion coating step. Essentially, the film die 10 is so positioned that the melt curtain 20, which is drawn down by gravity, will be deposited on the web 16 just at the point where the nip roller 12 and the quench roller 14 meet. This ensures that the pressure provided by the nip roller 12 is applied to the molten polymer resin straight after the polymer resin is extrusion coated onto the received web 16 of high-density paper 22. At the same time, the quench roller 14 is providing cooling of the coated substrate.

The precise polymer resin 24 that is to be used in the laminate structure 26 will depend on the ultimate application of the laminate structure 26. A

polyethylene polymer will be well suited to many applications, as such a polymer will provide the necessary barrier properties while also providing good heat sealing properties in the construction of a packaging container from the laminate structure 26. Such a polymer will also be readily and cheaply available and can be used in conventional extrusion coating machines. In more complex applications where specific additional barrier properties are needed from the laminate structure 26, then additional polymer resin layers can be extrusion coated onto the substrate, either before or after the extrusion coating of the polyethylene polymer.

Figure 5 shows a view similar to Figure 2 of the laminate structure 26, which comprises (in the simplest embodiment) a layer of high density paper 22 and a single layer of HDPE 24, which has been extrusion coated onto the high density paper 22 and then had pressure applied, as per the process of Figure 1. As detailed above, the surface properties of the treated high density paper 22 in combination with the readily crystallisable HDPE layer 24 (under pressure of between 2 and 20 bar) results in a portion 28 of the HDPE layer 24 achieving a higher level of crystallisation than the remainder of the layer 24.

For example, if the HDPE layer 24 is ten microns thick, then the external nine microns of HDPE will have a crystallisation of approximately 40%, whereas the portion 28 that is in contact with the high density paper 22 (at most one micron thick) will have a higher level of crystallisation, at least 55% and more likely in the 60% to 65% range. Higher crystallisation is achieved in this zone of the layer of readily crystallisable polyolefin polymer resin 24 through a combination of the physical properties of the polymer resin 24, the surface properties of the high density paper 22 and the application of pressure to the two layers 22 and 24.

The portion 28 of the polymer resin layer 24 that is more highly crystallised (at least 15% more crystallised) than the remainder of the polymer resin layer 24 is not in itself a separate layer. The result of the materials used and the process used is to produce the portion 28 within the layer 24 that has the higher level of crystallisation. This portion 28 provides the additional barrier properties without the need to use additional specific polymer resin layers. The

extrusion-coated structure 26 that combines the polymer layer 24 on the surface of the high-density paper 22 has a good oxygen barrier, which is achieved without the use of dedicated barrier resins.

CLAIMS

1. A method of producing a laminate structure, the method comprising the steps of:

- receiving a web of high-density paper formed by using refined chemical pulp and by subsequently treating the paper surface with alginic acid and/or starch and/or carboxyl methylcellulose and/or mechanical super calendaring and comprising a thickness of between 35 and 80 micrometre, a density of between 0.80 and 1.00 gr/cm^3 and a weight of between 30 and 80 gr/m^2,
- extrusion coating a layer of readily crystallisable polyolefin polymer resin onto the web of high-density paper, and
- applying a pressure of between 2 and 20 bar onto the layer of polyolefin polymer resin coated high-density paper thereby inducing crystallization in the portion of the polyolefin polymer resin adjacent to the high density paper, producing a level of crystallinity which is at least 15% higher than the remainder of the polyolefin polymer resin.

2. A method according to claim 1, and further comprising extrusion coating a second layer of polyolefin polymer resin onto the opposite side of the web of high-density paper as the first layer of polyolefin polymer resin.

3. A method according to claim 1 or 2, wherein the polyolefin polymer resin layer comprises a HDPE (high density polyethylene), a LDPE (low density polyethylene), a MDPE (medium density polyethylene), a LLDPE (linear low density polyethylene), a LMDPE (linear medium density polyethylene), a POP (polyolefin Plastomer), an EVA (ethylene vinyl acetate), an EMA (ethylene methyl acrylate), an EEA (ethylene ethyl acrylate), an EBA (ethylene butyl acrylate), an EAA (ethylene acrylic acid), or a zinc-, sodium- or lithium- based ionomer resin or a mix or blend of such polymers.

4. A method according to claim 1, 2 or 3, wherein the highly crystalline portion of the polyolefin polymer resin layer comprises a thickness of less than one micrometre.

5. A method according to any preceding claim, wherein the step of applying pressure onto the polyolefin polymer resin coated high-density paper occurs directly after the extrusion coating step.

6. A laminate structure comprising:

- a substrate of high-density paper, formed by using refined chemical pulp and by subsequently treating the paper surface with alginic acid and/or starch and/or carboxyl methylcellulose and/or mechanical super calendaring and comprising a thickness of between 35 and 80 micrometre, a density of between 0.80 and 1.00 gr/cm^3 and a weight of between 30 and 80 gr/m^2, and
- a layer of readily crystallisable polyolefin polymer resin that has been extrusion coated onto the web of high-density paper, the polyolefin polymer resin layer including crystallization in the portion of the polyolefin polymer resin adjacent to the high density paper, with a level of crystallinity which is at least 15% higher than the remainder of the polyolefin polymer resin.

7. A laminate structure according to claim 6, and further comprising a second layer of polyolefin polymer resin that has been extrusion coated onto the opposite side of the substrate of high-density paper as the first layer of polyolefin polymer resin.

8. A laminate structure according to claim 6 or 7, wherein the polyolefin polymer resin comprises a HDPE (high density polyethylene), a LDPE (low density polyethylene), a MDPE (medium density polyethylene), a LLDPE (linear low density polyethylene), a LMDPE (linear medium density polyethylene), a POP (polyolefin Plastomer), an EVA (ethylene vinyl acetate),

an EMA (ethylene methyl acrylate), an EEA (ethylene ethyl acrylate), an EBA (ethylene butyl acrylate), an EAA (ethylene acrylic acid), or a zinc-, sodium- or lithium- based ionomer resin or a mix or blend of such polymers.

9. A laminate structure according to claim 6, 7 or 8, wherein the highly crystalline portion of the polyolefin polymer resin layer comprises a thickness of less than one micrometre.

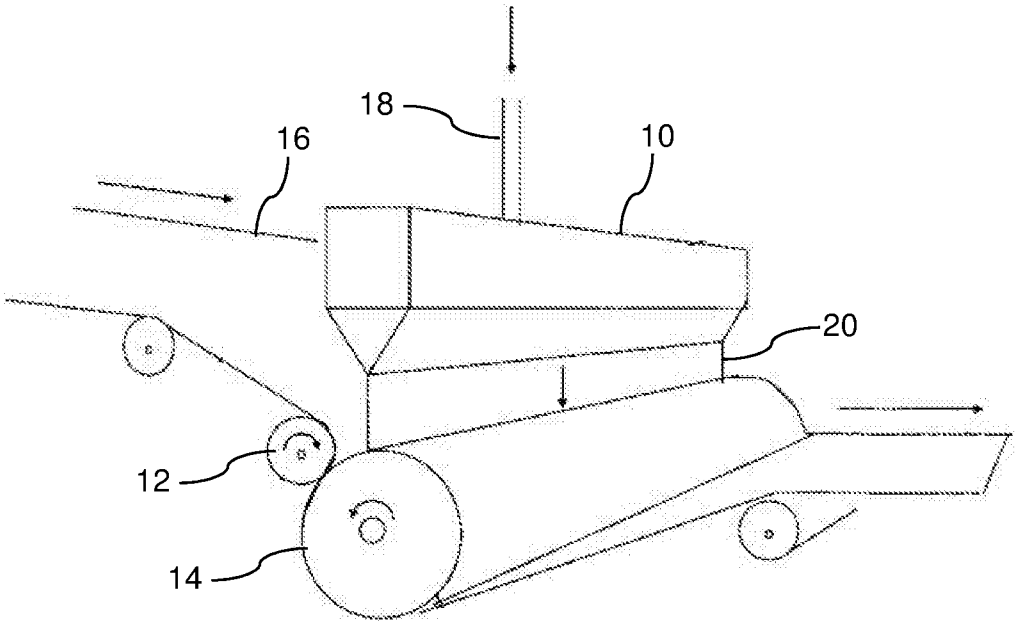


Fig. 1

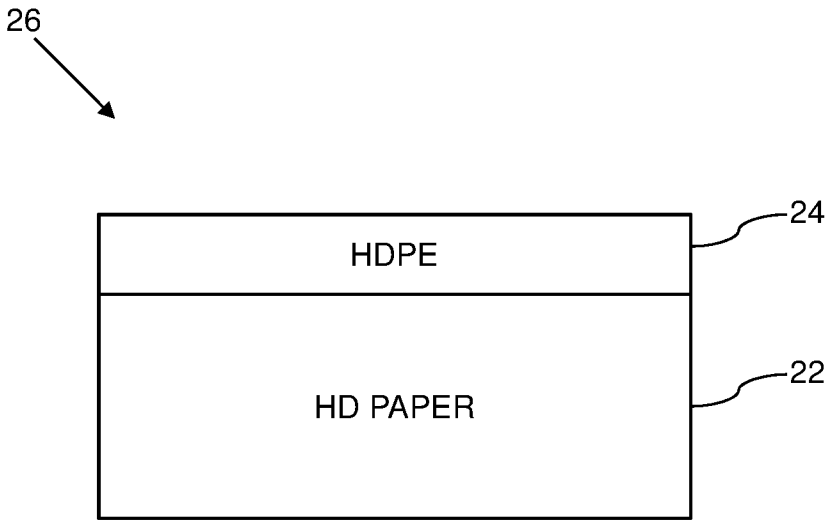


Fig. 2

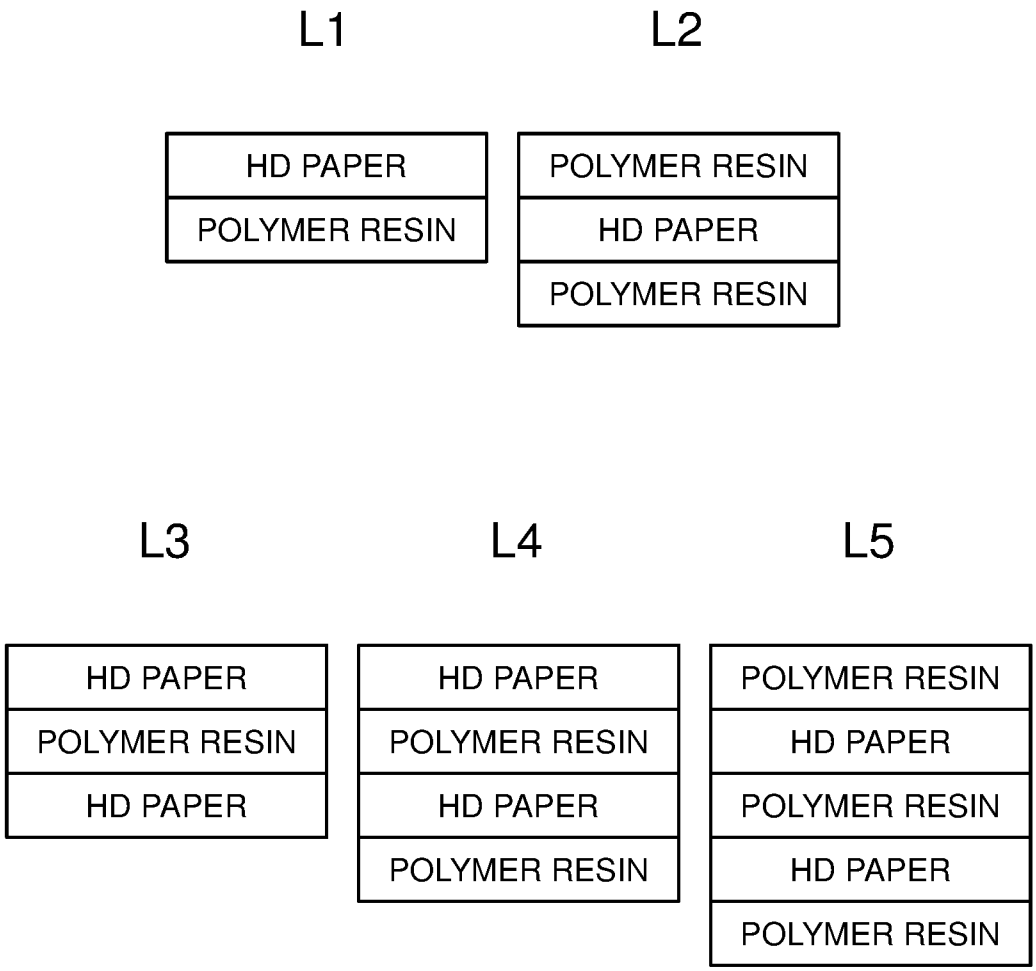

L1
L2
HD PAPER
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Fig. 3

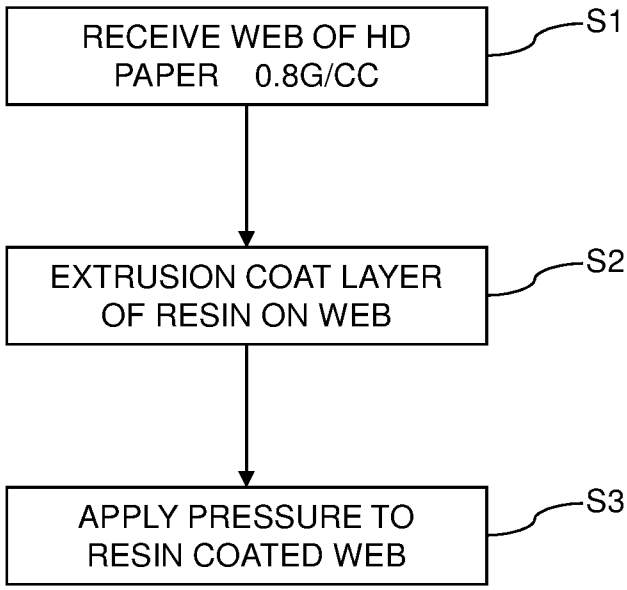

RECEIVE WEB OF HD PAPER 0.8G/CC
S1
EXTRUSION COAT LAYER OF RESIN ON WEB
S2
APPLY PRESSURE TO RESIN COATED WEB
S3

Fig. 4

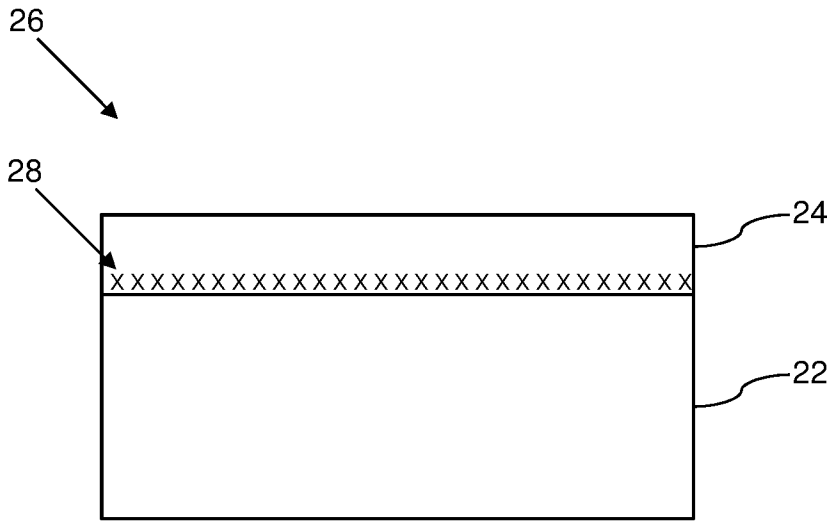


Fig. 5

INTERNATIONAL SEARCH REPORT

International application No
PCT/IB2014/061078

A. CLASSIFICATION OF SUBJECT MATTER
INV. B32B27/10 B32B27/30 B32B27/32
ADD.

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)
B32B D21H

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)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2005/053955 A1 (COMMW SCIENT IND RES ORG [AU]; STELLAR DEVELOPMENTS PTY LTD [AU]; GIBB) 16 June 2005 (2005-06-16) page 1, lines 4-8 page 11, line 10 - page 12, line 24 page 13, line 14 - page 15, line 27 page 17, line 9 - page 18, line 17 page 18, line 34 - page 19, line 36; figures 1-14 -----	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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Date of the actual completion of the international search	Date of mailing of the international search report
11 July 2014	21/07/2014

Name and mailing address of the ISA/	Authorized officer
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/IB2014/061078

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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