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(54) Title: BIOBASED ADHESIVE MIXTURE AND THE USE OF SAID ADHESIVE MIXTURE

(57) Abstract: The present invention relates to adhesive mixtures. In particular, the invention concerns the use of lignin in biobased hot-melt (non-pressure sensitive) and pressure sensitive adhesives, as well as novel adhesive mixtures suitable for use as such adhesives. The adhesive mixture comprises 25-85 wt-% lignin, 10-75 wt-% plasticizer, 0-50 wt-% tackifier and 0-40 wt-% waxes, by solid weight. The combined amount of lignin, plasticizer, tackifier and waxes is at least 90 wt-%, by solid weight, of the adhesive mixture.



BIOBASED ADHESIVE MIXTURE AND THE USE OF SAID ADHESIVE MIXTURE

Field of the invention

The present invention relates to adhesive mixtures. In particular, the invention
5 concerns the use of lignin in biobased hot-melt (non-pressure sensitive) and
pressure sensitive adhesives, as well as novel adhesive mixtures suitable for use as
such adhesives.

The adhesive composition can for example be used in packaging, for carton board,
10 textile, metal, glass, wood etc. as well as labels, where the adhesive can provide
either permanent or removable attachment. Other applications include wood
working, assembly, book binding and tapes.

Background

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Hot-melt adhesives are common for example in packaging for take-away products,
lamination for tissue paper and textiles as well as in boxes made of bio-based
materials.

20 Lignin, an aromatic polymer, is a major constituent in e.g. wood, being the most
abundant carbon source on Earth second only to cellulose. In recent years, with
development and commercialization of technologies to extract lignin in a highly
purified, solid and particularized form from the pulp-making process, it has attracted
significant attention as a possible renewable substitute to primarily aromatic
25 chemical precursors currently sourced from the petrochemical industry.

Lignin, being a polyaromatic network, has been extensively investigated as a suitable substitute for phenol during production of phenol-formaldehyde adhesives. These are used during manufacturing of structural wood products such as plywood, oriented strand board and fiberboard. During synthesis of such adhesives, phenol, partially/all replaced by lignin, is reacted with formaldehyde in the presence of either basic or acidic catalyst to form a highly cross-linked aromatic resins termed novolacs (when utilizing acidic catalysts) or resoles (when utilizing basic catalysts).

Hot melt adhesives (HMA) are a form of thermoplastic adhesive that are liquid when hot and then solidify in a period of from a few seconds to a few minutes, when cooled. Pressure-sensitive adhesives (PSAs) are adhesives that typically adhere to substrates when pressure is applied. The glue seam that is formed is normally not permanent, and the substrate is removable.

The pressure-sensitive adhesives (PSAs) are a sub-group of hot-melt adhesives, i.e. high-viscosity PSA mixtures that are heated to reduce viscosity enough to allow application onto a substrate, and subsequently they are cooled to their final form. They are thus capable of being applied as dispersions, solutions or hot melts, and subsequently give rise to a rubbery, tacky film of relatively low adhesive strength and higher cohesive strength at the service temperature.

Typically, pressure-sensitive adhesives can be used to produce bonds that are permanent, but not creep resistant. The PSAs can also be used to give rise to temporary or serial temporary bonds.

HMA (hot-melt adhesives) are available in a variety of different types, allowing for use in a wide range of applications across several industries. For use in industrial processes, the adhesive is typically supplied in larger sticks for use in glue guns with high melting rates. Aside from hot melt sticks, HMA can be delivered in other formats, such as granular, powder, pellet, pillow, block or cake for bulk melt processors. Larger applications of HMA traditionally use pneumatic systems to supply adhesive. Examples of industries where HMAs are used include: carton sealing and labeling applications in the packaging industry, spine gluing in the bookbinding industry, profile-wrapping, product assembly and laminating applications

in the woodworking industry, disposable diapers are constructed through the use of HMA, bonding the non- woven material to both the backsheet and the elastics, many electronic device manufacturers may also use an HMA to affix parts and wires, or to secure, insulate, and protect the device's components, and HMA are regularly used
5 to assemble and seal corrugated boxes and paperboard cartons. When applied upon a surface or into an interface, they give rise to a solid structure that is load-bearing at temperatures at which the treated surface or interface is being used (also called the operational temperature or the service temperature) .

10 In industrial use, hot-melt adhesives provide several advantages over solvent- based adhesives. Volatile organic compounds are reduced or eliminated, and the drying or curing step is eliminated. Hot melt adhesives have a long shelf life and can usually be disposed of without special precautions. HMAs also maintain their thickness when solidifying. Hot melt adhesives are useful for example in the environmentally
15 sensitive areas of packaging.

Lignin is non-linear phenolic biopolymer with rather low molar mass, and thus generally not considered suitable for use as a strength providing cohesive polymer for use in hot-melt adhesives.

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WO2018/122470 A1 discloses the use of lignins as tackifiers or waxy components in biobased hot-melt or pressure sensitive adhesives. The compositions also comprise hydrophobic cohesive polymers, such as EVA copolymers or polyolefins.

25 DE102012207868A1 discloses pressure-sensitive adhesive compositions comprising up to 20% by weight of lignin.

Prior art adhesive compositions comprising lignin often suffer from problems arising from inhomogeneous polymer blends.

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There is a need to provide hot-melt formulations with a high proportion of renewable materials, ideally fully biobased and/or biodegradable, but with adequate flow properties and binding strength.

5 Summary of the invention

Kraft lignin has a high glass transition temperature and does not flow well at elevated temperatures, not even above its glass transition temperature. However, it has been found that the lignin melting behaviour can be improved by adding plasticizers such as triacetin, triethyl citrate, propylene carbonates, ethylene carbonate, polyethylene glycol, polypropylene glycol, ethylene glycol or propylene glycol. The type of the plasticizers and their amount controls the lignin melt viscosity and softening point of the adhesive formulations.

The bonding strength of the molten lignin normally requires another cohesive polymer. It was surprisingly found that a formulation comprising lignin, biobased plasticizers, optionally tackifiers (non-biobased or biobased, mainly based on rosins or terpenes) and optionally waxes (preferably biobased), in the lignin hot-melt formulations significantly improves the bonding strength of the adhesives.

Surprisingly, it was found that hot melt adhesive formulations according to the present invention showed good performance in the packaging applications without requiring any another hydrophobic polymer other than lignin.

The present invention is thus directed to an adhesive mixture comprising 25-85 wt-% lignin, 10-75 wt-% plasticizer, 0-50 wt-% tackifier and 0-40 wt-% waxes, by solid weight, wherein the combined amount of lignin, plasticizer, tackifier and waxes is at least 90 wt-%, by solid weight, of the adhesive mixture.

The present invention is also directed to a method for preparing the adhesive mixture.

The present invention is thus also directed to the use of the adhesive mixture in carton sealing and labeling, paperboard assembling and sealing, spine gluing in the bookbinding industry, profile-wrapping, product assembly and laminating applications in the woodworking industry, installation of flooring and ceiling panels, gluing of
5 woven and non-woven fabrics, disposable diapers, affixing of parts and wires in electronic devices, or to secure, insulate, and protect the device's components.

Detailed description

10 It is intended throughout the present description that the expression "lignin" embraces any kind of lignin, e.g. lignin originated from hardwood, softwood or annular plants. Preferably the lignin is an alkaline lignin generated in e.g. the Kraft process. Preferably, the lignin has been purified or isolated before being used in the process according to the present invention. The lignin may be isolated from black
15 liquor and optionally be further purified before being used in the process according to the present invention. The purification is typically such that the purity of the lignin is at least 90%, preferably at least 95%, more preferably at least 98% or 99%. Thus, the lignin used according to the method of the present invention preferably contains less than 10%, preferably less than 5%, more preferably less than 2% or 1%
20 impurities. The lignin may then be separated from the black liquor by using the process disclosed in WO2006031175. The lignin may then be separated from the black liquor by using the process referred to as the LignoBoost process. The lignin may be provided in the form of particles, such as particles having an average particle size of from 50 micrometers to 500 micrometers. The lignin may be chemically
25 modified using methods known in the art. Preferably, the lignin used according to the present invention has not been chemically modified.

"Plasticizers" are components that increase the fluidity of mixtures and compositions and make them softer. "External" plasticizers are separate components added to the
30 adhesive mixtures to increase their plasticity, while "internal" plasticizers are inherent parts of the polymer molecules and become part of the product. Internal plasticizers can be either co-polymerized into the polymer structure or reacted with the original polymer.

Any known plasticizers can be used, preferred examples of polymeric plasticizers thereof include the following: polyethylene glycol, polypropylene glycol, polyglycerol castor oil, olive oil, rapeseed oil, pine oil and mixtures thereof. Preferred examples of
5 monomeric plasticizers include the following: ethylene glycol, propylene glycol, glycerol, propylene carbonate, ethylene carbonate, glycerol carbonate, vinyl carbonate, triacetin, diacetin, monoacetin, triethyl citrate, tributyl citrate, acetyl triethyl citrate, acetyl tributyl citrate, dimethyl succinate, diethyl succinate, ethyl lactate, methyl lactate, fatty acid esters of glycerol or polyglycerol (such as
10 polyglycerol caprylate) or polyethylene glycol, diethyl phthalate, and mixtures thereof. Polyglycerol is one example suitable for reducing the T_g (glass transition temperature) as well as the viscosity of the adhesive mixture. Its content in these mixtures is typically from about 10 to about 75 wt-% by weight of the mixture, based on the dry weight. Preferably, the plasticizers used are polymeric plasticizers or bio-
15 based plasticizer or more preferably, bio-based polymeric plasticizers. In one embodiment, the plasticizer is a fatty acid ester of polyglycerol, such as polyglycerol caprylate.

The term "tackifier", refers to the component of an adhesive mixture that provides the
20 adhesive with external strength, i.e. that is capable of holding the adhesive and the substrate together. The tackifier may be synthetic or biobased. Preferably, the tackifier is a biobased tackifier, such as a tackifier mainly based on rosins or terpenes, such as rosin acid, rosin ester, hydrogenated rosin ester, terpene phenolic resin etc.

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The adhesive mixture according to the present invention optionally comprises waxes, e.g., synthetic waxes, bees wax, vegetable oil-based wax, fatty amide waxes or oxidized Fischer-Tropsch waxes. Preferably, the wax is bio-based wax such as vegetable-oil based wax, soy-based wax or bees wax etc. These waxes function by
30 increasing the setting rate, and by lowering the melt viscosity. Further, they can improve bond strength and temperature resistance.

The adhesives (also called glues) of the present invention include hot-melt adhesives (HMA), which are soft and tacky when hot, and solidify upon cooling, as well as pressure-sensitive adhesives (PSA), which adhere to substrates when pressure is applied.

5

The adhesive mixture according to the present invention optionally comprises extenders such as antioxidants and stabilizers (e.g., hindered phenols, butylated hydroxytoluene (BHT), phosphites, phosphates, hindered aromatic amines), added in small amounts (<1 wt-%), not influencing physical properties, but protecting the material from degradation or ageing (caused, e.g., by autoxidation), or UV stabilizers, or biocides for hindering bacterial growth, or flame retardants.

10

The adhesive mixture according to the present invention optionally comprises pigments, dyes and/or glitter.

15

The adhesive mixture according to the present invention may optionally comprise antistatic agents. The adhesive mixture according to the present invention may optionally comprise organic carboxylic acids having at least two carboxyl groups, such as citric acid. The adhesive mixture according to the present invention may optionally comprise agents that make the mixture more hydrophobic, such as cellulose acetate.

20

Preferably, the adhesive mixture comprises 25-55 wt-% lignin, 20-65 wt-% plasticizer, 0-35 wt-% tackifier and 0-5 wt-% waxes, by solid weight.

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More preferably, the adhesive mixture comprises 40-55 wt-% lignin, 25-40 wt-% plasticizer, 15-25 wt-% tackifier and 0-5 wt-% waxes, by solid weight.

The combined amount of lignin, plasticizer, tackifier and waxes is at least 90 wt-%, by solid weight, of the adhesive mixture. Preferably, the combined amount of lignin,

30

plasticizer, tackifier and waxes is at least 92 wt-%, more preferably at least 95 wt-%, by solid weight, of the adhesive mixture.

5 The adhesive mixture may be prepared by mixing the selected components in the desired ratios at an elevated temperature, particularly varying from about 60° to about 200°C, preferably at a temperature of from 100° to 150°C.

10 In one embodiment, the adhesive mixture may be prepared stepwise. In a first step, lignin is added to the plasticizer, optionally in the presence of tackifier and/or wax at a temperature of from 50° to 160°C. In a second step, the dispersion obtained in the first step is heated to a temperature of from 100° to 235°C, to produce a homogenous adhesive mixture without creating any lumps.

15 In general, hot-melt adhesive mixtures are applied to the selected substrates by jet application in various forms (extrusion, spray, slot, spot). The high melt viscosity makes them ideal for porous and permeable substrates. HMAs are capable of bonding an array of different substrates including: rubbers, ceramics, metals, plastics, glass and wood.

20 According to an embodiment of the present invention, the prepared adhesive mixture is applied and glued on a suitable substrate selected from rubbers, ceramics, metals, plastics, glass, wood, paper and board substrates, with paper and board substrates being the most suitable alternatives, such as sack paper and coated board.

25 After application, the important gluing parameters in industrial processes are the open time related to the setting behavior, and compression phase (close time/pressure).

30 According to a preferred embodiment, elevated temperature and pressure are used during the application of the adhesive mixture. Examples of suitable parameters include an application temperature of 100° to 200°C, preferably 120° to 190°C, and most suitably about 120° to 170°C.

The adhesives of the present invention can be used in a wide variety of applications,

- e.g., carton sealing and labeling, paperboard assembling and sealing, spine gluing in the bookbinding industry, profile-wrapping, product assembly and laminating applications in the woodworking industry, installation of flooring and ceiling panels, gluing of woven and non-woven fabrics, disposable diapers, affixing of parts and
- 5 wires in electronic devices, or to secure, insulate, and protect the device's components.

Examples

Example 1

- 10 Lignin powder (solid content 95%), polyglycerol-4 and vegetable wax was mixed in a weight ratio of 100:150:33 in a 400ml glass beaker, the beaker was then immersed in the heated oil bath on a hot plate, the mixture was melt compounded (hot blended) for 4 hours at 160°C under over-head stirring using 3 blade propeller central shaft. The homogeneous molten product was formed visually, the product made was able
- 15 to solidify upon cooling and turned to the molten state again when undergo reheating.

Example 2

- Lignin powder (solid content 95%), polyglycerol-4 and rosin gum was mixed in a
- 20 weight ratio of 100:150:33 in a 400ml glass beaker, the beaker was then immersed in the heated oil bath on a hot plate, the mixture was melt compounded (hot blended) for 4 hours at 160°C under over-head stirring using 3 blade propeller central shaft. The homogeneous molten product was formed visually, the product made was able to solidify upon cooling and turned to the molten state again when undergo
- 25 reheating.

Example 3

- Granulated lignin powder (solid content 98%), polyglycerol-4 and vegetable wax was mixed in a weight ratio of 100:150:33 in a 400ml glass beaker, the beaker was then
- 30 immersed in the heated oil bath on a hot plate, the mixture was melt compounded (hot blended) for 4 hours at 160°C under over-head stirring using 3 blade propeller central shaft. The homogeneous molten product was formed visually, the product

made was able to solidify upon cooling and turned to the molten state again when undergo reheating.

Example 4

- 5 Lignin powder (solid content 95%), polyglycerol-4 and PEG 400 was mixed in a weight ratio of 100:33:33 in a 400ml glass beaker, the beaker was then immersed in the heated oil bath on a hot plate, the mixture was melt compounded (hot blended) for 4 hours at 160°C under over-head stirring using 3 blade propeller central shaft. The homogeneous molten product was formed visually, the product made was able
10 to solidify upon cooling and turned to the molten state again when undergo reheating.

Example 5

- Lignin powder (solid content 95%), triethyl citrate, citric acid was hand mixed 5 min in
15 a glass beaker with weight ratio of 50:41:10, the beaker was then baked in the oven at 160°C for 20 min. The homogeneous molten product was formed visually, the product made was able to solidify upon cooling and turned to the molten state again when undergo reheating.

Example 6

- Lignin powder (solid content 95%), triacetin, 2,5-furandicarboxylic acid was hand mixed 5 min in a glass beaker with weight ratio of 50:41:10, the beaker was then baked in the oven at 160°C for 20 min. The homogeneous molten product was formed visually, the product made was able to solidify upon cooling and turned to the
25 molten state again when undergo reheating.

Example 7

- 41g Polyglycerol-4 and 41g PEG 400 were mixed in a 400ml glass beaker, the beaker was then immersed in an oil bath had reached 120°C on the hot plate. After
30 the immersion, the mixture was kept at 120°C for 20 min under mild over-head stirring (150 rpm) using 3 blade propeller central shaft. This warmup step was evidenced transforming the sticky semi-solid polyglycerol-4, which sinks at the

bottom of the beaker to an easy flowable liquid which significantly eased the lignin addition afterwards. 100g lignin powder (solid content 95%) were then added in small portions manually to this pre-warmed mixture over a period of about 15 min, with rapid stirring at 350 rpm. After the adding was complete, the dispersion of lignin
5 was continued for another 30 min. After the dispersion step, the heating temperature was set to rapidly ramp up to 150°C. The homogeneous molten product was formed visually in an hour with mild stirring at 180 rpm. the product made was able to solidify upon cooling and turned to the molten state again when undergo reheating.

10 Application and testing

ABES, the automated bonding evaluation system was used to determine the dry shear strength on a lap shear test. The test sample, D flute (2mm) corrugated cardboard was cut to 20 mm wide and 105 mm in length. For each test, two corrugated strips were glued together with an overlapping joint area of 20×5 mm².
15 The glue was added to the tank of the hand-held applicator either in its solid form or at its molten state. 45 min stabilization time was allowed for the hand-held applicator to reach the operating temperature at 150°C. By pulling back on the trigger of the applicator, the glue was carefully dispensed along the area mark line on the first substrate, and the second substrate was rapidly closed aligning to their
20 corresponding bonding area, any excess glue will be squeezed out from the marked joint area during immediately 3 seconds hand pressing. Tests were performed by 4 seconds pulling after one day lab conditioning, the average data from 5 test specimens was shown in the table below.

Sample ID	Dry shear strength N/ mm ²	Adhesive failure
Example 1	1.4	100% substrate
Example 2	1.8	100% substrate
Example 3	2.5	100% substrate
Example 4	1.7	100% substrate
Example 5	2.8	100% substrate
Example 6	2.9	100% substrate
Example 7	2.2	100% substrate
Technomelt supra 100 (Henkel) (reference)	2.5	100% substrate

In view of the above detailed description of the present invention, other modifications
5 and variations will become apparent to those skilled in the art. However, it should be
apparent that such other modifications and variations may be effected without
departing from the spirit and scope of the invention.

Claims

- 5 1. An adhesive mixture comprising 25-85 wt-% lignin, 10-75 wt-% plasticizer, 0-50 wt-% tackifier and 0-40 wt-% waxes, by solid weight, wherein the combined amount of lignin, plasticizer, tackifier and waxes is at least 90 wt-%, by solid weight, of the adhesive mixture.
- 10 2. An adhesive mixture according to claim 1, which is in the form of hot-melt adhesive formulation.
- 15 3. An adhesive mixture according to claim 1 or 2, comprising 40-55 wt-% lignin, 25-50 wt-% plasticizer, 15-35 wt-% tackifier and 0-5 wt-% waxes, by solid weight.
4. An adhesive mixture according to claim 3, comprising 40-55 wt-% lignin, 25-40 wt-% plasticizer, 15-25 wt-% tackifier and 0-5 wt-% waxes, by solid weight.
- 20 5. An adhesive mixture according to any one of claims 1-4, wherein the plasticizer is polyglycerol, polyethylene glycol or polypropylene glycol or a mixture thereof.
- 25 6. An adhesive mixture according to any one of claims 1-5, wherein the tackifier is based on rosins or terpenes.
- 30 7. An adhesive mixture according to any one of claims 1-6, wherein the purity of the lignin is at least 98% and the lignin has not been chemically modified.

8. A method for preparing an adhesive mixture according to any one of claims 1-7, comprising the step of mixing the lignin, plasticizer, tackifier and optionally waxes at an elevated temperature of from 100° to 200°C.
- 5 9. Use of an adhesive mixture according to any one of claims 1-7 in carton sealing and labeling, paperboard assembling and sealing, spine gluing in the bookbinding industry, profile-wrapping, product assembly and laminating applications in the woodworking industry, installation of flooring and ceiling panels, gluing of woven and non-woven fabrics,
- 10 disposable diapers, affixing of parts and wires in electronic devices

INTERNATIONAL SEARCH REPORT

 International application No.
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A. CLASSIFICATION OF SUBJECT MATTER IPC: see extra sheet According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) IPC: C08L, C09J		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched SE, DK, FI, NO classes as above		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) EPO-Internal, PAJ, WPI data, polyindex		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 2011219728 A (HITACHI CHEMICAL CO LTD), 4 November 2011 (2011-11-04); abstract; paragraphs [0001], [0020], [0026]-[0030], [0033], [0044] --	1-9
X	US 20110054154 A1 (PARK DAVID W ET AL), 3 March 2011 (2011-03-03); paragraphs [0001], [0008]-[0032]; all claims --	1-9
X	EP 3632866 A1 (ROCKWOOL INT A/S), 8 April 2020 (2020-04-08); paragraphs [0013]-[0017], [0020]-[0024], [0057]-[0064], [0194]-[0196] --	1, 5, 7, 9
A	--	2-4, 6, 8
☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.		
*	Special categories of cited documents:	
“A”	document defining the general state of the art which is not considered to be of particular relevance	“T” later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
“D”	document cited by the applicant in the international application	“X” document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
“E”	earlier application or patent but published on or after the international filing date	
“L”	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	“Y” document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
“O”	document referring to an oral disclosure, use, exhibition or other means	
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Date of the actual completion of the international search	Date of mailing of the international search report	
13-05-2022	13-05-2022	
Name and mailing address of the ISA/SE Patent- och registreringsverket Box 5055 S-102 42 STOCKHOLM Facsimile No. + 46 8 666 02 86	Authorized officer Ingemar Wistrand Telephone No. + 46 8 782 28 00	

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 International application No.
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C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	EP 0634466 A2 (NAT STARCH CHEM INVEST), 18 January 1995 (1995-01-18); abstract; page 2, line 1 - page 2, line 35; page 3, line 45 - page 4, line 55; all claims	1-9
A	US 4702496 A (HUME III ROBERT M), 27 October 1987 (1987-10-27); column 11, line 3 - line 45; column 13, line 46 - column 14, line 34; claims 1,3,6	1-9
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INTERNATIONAL SEARCH REPORT

Information on patent family members

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