



FI000123934B

(12) **PATENTTIJULKAISU**
PATENTSKRIFT(10) **FI 123934 B**

(45) Patentti myönnetty - Patent beviljats

31.12.2013

(51) Kv.lk. - Int.kl.

C08H 7/00 (2011.01)
C07G 1/00 (2011.01)
C08G 8/24 (2006.01)
C08L 97/00 (2006.01)**SUOMI – FINLAND****(FI)****PATENTTI- JA REKISTERIHALLITUS**
PATENT- OCH REGISTERSTYRELSEN

(21) Patentihakemus - Patentansökning

20125357

(22) Saapumispäivä - Ankomstdag

29.03.2012

(24) Tekemispäivä - Ingivningsdag

29.03.2012

(41) Tullut julkiseksi - Blivit offentlig

30.09.2013

(73) Haltija - Innehavare

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Matalamolekyylipainoisen ligniinin käyttö sideainekoostumuksen valmistamiseksi
Användning av lignin med låg molekylvikt för framställning av en bindemedelskomposition

(56) Viitejulkaisut - Anförda publikationer

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(57) Tiivistelmä - Sammandrag

The present invention relates to a method for producing a binder composition, wherein the method comprises the following steps: (i) forming an aqueous composition comprising reactant components including high molecular weight lignin, low molecular weight lignin, polymerizable substance and crosslinking agent in the presence of a catalyst; and (ii) cooking the composition at a temperature of 60 - 95 °C for polymerizing the reactant components until a binder composition with a predetermined viscosity value is formed; wherein the high molecular weight lignin comprises lignin molecules of 11 - 60 lignin units, and wherein the low molecular weight lignin comprises lignin molecules of 1 - 10 lignin units.

Esillä oleva keksintö koskee menetelmää sideainekoostumuksen valmistamiseksi, jossa menetelmä käsittää seuraavat vaiheet: (i) muodostetaan vesipitoinen koostumus, joka käsittää reagenssikomponentteja, jotka sisältävät suurimolekyylipainoista ligniiniä, pienimolekyylipainoista ligniiniä, polymerisoituvaa ainetta ja silloittavaa ainetta, katalyytin läsnä ollessa; ja (ii) keitetään koostumusta 60 - 95 °C:n lämpötilassa reagenssikomponenttien polymerisoimiseksi, kunnes muodostuu sideainekoostumus, jolla on ennalta määrätty viskositeetti-arvo; jossa suurimolekyylipainoinen ligniini käsittää 11 - 60 ligniiniyksikön ligniinimolekyylejä ja jossa pienimolekyylipainoinen ligniini käsittää 1 - 10 ligniiniyksikön ligniinimolekyylejä.

USE OF LOW MOLECULAR WEIGHT LIGNIN FOR THE PRODUCTION
OF A BINDER COMPOSITION

FIELD OF THE INVENTION

5 The invention relates to a method for producing a binder composition, to a binder composition and to different applications of the binder composition.

BACKGROUND OF THE INVENTION

10 Lignin is a natural polymer, which can be extracted from e.g wood. As lignin is a natural biopolymer its use as a component in glues instead of synthetic materials has been investigated in order to come up with a more environmentally friendly adhesive
15 composition. Especially, the ability to replace synthetic phenol in phenolic resins, such as phenol formaldehyde resin, has been the object of prior art.

 Different types of adhesive compositions, such a phenolic glues, can be used with wood products.
20 Examples of such glues include compositions comprising phenol formaldehyde resin. Traditionally synthetic phenol formaldehyde resins are produced by polymerizing phenol and formaldehyde in the presence of a catalyst. Examples of such catalysts are sodium hydroxide
25 (NaOH) and acids. The method for producing phenol formaldehyde resin comprises adding formaldehyde in a stepwise manner to a phenol composition and thereafter increasing the temperature of the formed composition up to 80 - 90 °C. The composition is cooked at this
30 temperature until a desired viscosity of the formed resin or polymer chain length is reached.

 Lignin can be used for the purpose of decreasing the amount of synthetic phenol in a resin composition. Lignin has previously been used for replacing phenol during the production of lignin-phenol-
35 formaldehyde resin.

It has been possible to replace up to 30 % of the synthetic phenol in the final resin, e.g. phenol formaldehyde resin, with lignin but higher replacement has resulted in unsatisfying properties of the produced glue.

The inventors have therefore recognized a need for a method, which would result in a higher phenol replacement in the composition and thus in a more environmentally friendly binder composition having suitable properties for use in different applications.

PURPOSE OF THE INVENTION

The purpose of the invention is to provide a new type of method for replacing at least part of the amount of synthetic materials used during the production of a binder composition. Especially the purpose is to produce a more environmentally friendly binder composition to be used e.g. in adhesive applications.

SUMMARY

The method according to the present invention is characterized by what is presented in claim 1.

BRIEF DESCRIPTION OF THE DRAWINGS

The accompanying drawings, which are included to provide a further understanding of the invention and constitutes a part of this specification, illustrate some embodiments of the invention and together with the description helps to explain the principles of the invention. In the drawings:

Fig. 1 is a flow chart illustration of a method according to one embodiment of the present invention; and

Fig. 2 is a flow chart illustration of a method according to another embodiment of the present invention.

5 DETAILED DESCRIPTION OF THE INVENTION

The present invention relates to a method for producing a binder composition, wherein the method comprises the following steps:

10 (i) forming an aqueous composition comprising reactant components including high molecular weight lignin, low molecular weight lignin, polymerizable substance and crosslinking agent in the presence of a catalyst; and

15 (ii) cooking the composition at a temperature of 45 - 95 °C for polymerizing the reactant components until a binder composition with a predetermined viscosity value is formed;

wherein the high molecular weight lignin comprises lignin molecules of 11 - 60 lignin units, and
20 wherein the low molecular weight lignin comprises lignin molecules of 1 - 10 lignin units.

The expression "high molecular weight lignin" should be understood in this specification, unless otherwise stated, as comprising lignin molecules of 11
25 - 60 lignin units.

The expression "low molecular weight lignin" should be understood in this specification, unless otherwise stated, as comprising lignin molecules of 1
- 10 lignin units..

30 The expression "lignin unit" should be understood in this specification, unless otherwise stated, as phenylpropane. The phenylpropane can comprise OH- and/or OMe-groups attached thereto. The lignin units are crosslinked to each other through ether linkages.
35 One phenylpropane comprises nine carbon atoms.

A drawback of different methods for separating or isolating lignin from e.g. biomass is that the

lignin is condensed during the procedure due to the low pH environment used. Thus, separated lignin has a rather low reactivity and a heterogenic nature, which affect the reactions with other reactant components during the production of a binder composition. The low reactivity of lignin has been one of the reasons preventing a higher replacement level of e.g. synthetic phenol in binder compositions with biobased lignin. It has been recognized that the properties of currently available binder compositions, wherein up to 50 - 60 % of the synthetic phenol has been replaced with lignin, are not acceptable for e.g. gluing applications. E.g. the strength of glued joints has not been on a required level.

The inventors surprisingly found out that by using low molecular weight lignin in combination with a fraction of high molecular weight lignin during the production of a binder composition a higher replacement level of e.g. synthetic phenol in binder compositions can be achieved. It was found out that the use of low molecular weight lignin, which is recognized as more reactive compared to high molecular weight lignin, e.g. boosts the reactions taking place during the binder composition production.

In one embodiment of the present invention the high molecular weight lignin is separated from black liquor, kraft lignin, biomass originating lignin, lignin from alkaline pulping process, lignin from soda process, lignin from organosolv pulping or from a combination thereof. In one embodiment of the present invention the low molecular weight lignin is separated from black liquor, kraft lignin, biomass originating lignin, lignin from alkaline pulping process, lignin from soda process, lignin from organosolv pulping or from a combination thereof.

Low molecular weight lignin as well as high molecular weight lignin can be separated from differ-

ent sources using different kinds of methods or procedures. Low molecular weight lignin can be separated from e.g. kraft lignin by ultrafiltration, nanofiltration or by sequential precipitation. Low molecular
5 lignin can also be separated from black liquor by e.g. ultrafiltration.

In this specification, unless otherwise stated, the expression "lignin" should be understood as any lignin suitable to be used in the present invention including essentially pure lignin.
10

By the expression "essentially pure lignin" should be understood as at least 90 % pure lignin, preferably at least 95 % pure lignin. In one embodiment of the present invention the essentially pure
15 lignin comprises at most 10 %, preferably at most 5 %, of other components. Extractives and carbohydrates such as hemicelluloses can be mentioned as examples of such other components.

By "kraft lignin" is to be understood in this specification, unless otherwise stated, lignin that originates from kraft black liquor. Black liquor is an alkaline aqueous solution of lignin residues, hemicellulose, and inorganic chemicals used in a kraft pulping process. The black liquor from the pulping process
20 comprises components originating from different softwood and hardwood species in various proportions. Lignin can be separated from the black liquor by different, techniques including e.g. precipitation and filtration. Lignin usually begins precipitating at pH
25 values below 11 - 12. Different pH values can be used in order to precipitate lignin fractions with different properties. These lignin fractions differ from each other by molecular weight distribution, e.g. Mw and Mn, polydispersity, hemicellulose and extractive
30 contents. The molar mass of lignin precipitated at a higher pH value is higher than the molar mass of lignin precipitated at a lower pH value. Further, the mo-
35

lecular weight distribution of lignin fraction precipitated at a lower pH value is wider than of lignin fraction precipitated at a higher pH value. Thus the properties of the lignin can be varied depending on
5 the end use of the gluing application.

The precipitated lignin can be purified from inorganic impurities, hemicellulose and wood extractives using acidic washing steps. Further purification can be achieved by filtration.

10 In one embodiment of the present invention the dry matter content of the lignin, e.g. the lignin, is below 98 %, preferably 40 - 80 %, and more preferably 50 - 70 %.

In one embodiment of the present invention
15 the lignin is separated from pure biomass. The separation process can begin with liquidizing the biomass with strong alkali followed by a neutralization process. After the alkali treatment the lignin can be precipitated in a similar manner as presented above.

20 In one embodiment of the present invention the separation of lignin from biomass comprises a step of enzyme treatment. The enzyme treatment modifies the lignin to be extracted from biomass. Lignin separated from pure biomass is sulphur-free and thus valuable in further
25 processing.

In one embodiment of the present invention the predetermined viscosity value of the final binder composition is at least 40 cP, preferably at least 50 cP, and more preferably at least 80 cP. In one embodi-
30 ment of the present invention the predetermined viscosity value of the final binder composition is at least 40 but not more than 250 cP, preferably at least 50 cP but not more than 150 cP, and more preferably at least 80 but not more than 120 cP.

35 In one embodiment of the present invention the predetermined viscosity value of the final binder composition is at least 250 cP, preferably at least

300 cP, and more preferably at least 500 cP. In one embodiment of the present invention the predetermined viscosity value of the final binder composition is at least 250 cP but not more than 1500 cP, preferably at least 300 cP but not more than 1200 cP, and more preferably at least 500 but not more than 1000 cP. The viscosity is measured at 25 °C using a rotary viscometer. The predetermined viscosity value of the final binder composition may vary depending on the specific application where the binder composition is to be used.

In one embodiment of the present invention the high molecular weight lignin is alkalated before being used in the method for producing a binder composition. In one embodiment of the present invention the low molecular weight lignin is alkalated before being used in the method for producing a binder composition. Alkalation of lignin makes the lignin more reactive. In one embodiment the alkalation comprises forming, under heating at a temperature of 30 - 70 °C, an aqueous dispersion comprising alkali and lignin, and heating the formed dispersion at a temperature of 50 - 95 °C for 15 minutes - 2 hours for producing alkalated lignin. The alkali may comprise a hydroxide of an alkali metal. Treating the lignin by alkalation activates the lignin making it more suitable for use in further applications.

In one embodiment of the present invention the weight ratio of high molecular weight lignin to low molecular weight lignin is 0,1 - 20, preferably 0,2 - 10, and more preferably 0,5 - 5.

In one embodiment of the present invention step (i) of forming the aqueous composition comprises the following steps:

(ia) forming a dispersion comprising high molecular weight lignin; and

(ib) combining polymerizable substance and crosslinking agent with the dispersion for forming the aqueous composition.

In one embodiment of the present invention
5 step (ib) comprises adding catalyst.

In one embodiment of the present invention step (i) of forming the aqueous composition comprises, after step (ib), the following steps:

(ic) cooking the formed composition until the
10 composition has a viscosity, which corresponds to 15 - 70 % of a predetermined viscosity value of the final binder composition; and

(id) adding low molecular weight lignin as a reactant component to the composition.

In one embodiment of the present invention
15 step (ia) and/or step (ib) comprises adding low molecular weight lignin as a reactant component.

In one embodiment of the present invention step (ia) comprises dissolving lignin in a solvent. In
20 one embodiment of the present invention the solvent comprises sodium hydroxide. In one embodiment of the present invention step (ia) comprises adding water.

The temperature of the dispersion may increase during step (ia) from room temperature up to 70
25 °C.

In one embodiment all of the amount of the polymerizable substance, e.g. phenol, used is added at once into the composition. In one embodiment of the present invention the crosslinking agent, e.g. aldehyde,
30 hyde, is added in a stepwise manner. Adding the aldehyde in a stepwise manner ensures that the temperature of the composition is not increased too much or too quickly.

In one embodiment of the present invention
35 step (i) comprises keeping the composition at a temperature of at most 75 °C.

In one embodiment of the present invention step (ic) comprises keeping the temperature of the composition between 40 - 95 °C, preferably between 50 - 85 °C, and more preferably between 60 - 80 °C.

5 In one embodiment of the present invention the temperature of the composition is decreased below 85 °C, and preferably below 65 °C before step (id).

In one embodiment of the present invention step (ii) comprises cooking the composition preferably
10 at a temperature of 50 - 85°C, and more preferably at a temperature of 60 - 80 °C.

In one embodiment of the present invention the temperature of the composition is increased up to 45 - 95 °C, preferably up to 50 - 85 °C, and more preferably up to 60 - 80 °C, before, during or after step
15 (id).

The temperature can be controlled during the production of the binder composition by cooling and/or heating the composition.

20 In one embodiment of the present invention the weight ratio of high molecular weight lignin and low molecular weight lignin to the polymerizable substance is at least 0,2, preferably at least 1, and more preferably at least 5.

25 In one embodiment of the present invention the amount of low molecular weight lignin is 1 - 95 %, preferably 3 - 60 %, and more preferably 5 - 30 %, of the amount of high molecular weight lignin.

In one embodiment of the present invention
30 the relation between the amounts of high molecular weight lignin, catalyst/solvent, polymerizable substance, crosslinking agent and low molecular weight lignin, based on their dry contents, used for producing the binder composition is the following: 18 - 60
35 weight-%, preferably 26 - 45 weight-%, of crosslinking agent and catalyst/solvent, and 82 - 40 weight-%, preferably 74 - 55 weight-%, of the polymerizable sub-

stance, high molecular weight lignin and low molecular weight lignin.

In one embodiment of the present invention the crosslinking agent is selected from a group consisting of an aldehyde, a derivative of an aldehyde, an aldehyde forming compound and combinations thereof. In one embodiment of the present invention the derivative of an aldehyde is hexamethylenetetramine, paraformaldehyde or trioxane. In one embodiment of the present invention the crosslinking agent is selected from a group consisting of an aromatic aldehyde, glyoxal, furfuryl alcohol, caprolactam and glycol compounds. The aldehyde can be formaldehyde. The aromatic aldehyde can be furfuryl aldehyde. In one embodiment of the present invention the crosslinking agent is a bio-based crosslinking agent. In one embodiment of the present invention the crosslinking agent is an aldehyde, and preferably formaldehyde.

In one embodiment of the present invention the polymerizable substance is a compound selected from the class of phenols. In one embodiment of the present invention the polymerizable substance is selected from a group consisting of phenol, cresol, resorcinol and combinations thereof. In one embodiment of the present invention the polymerizable substance is phenol. In one embodiment of the present invention the polymerizable substance is selected from a group consisting of bio-based hydroxyphenols and their derivatives. In one embodiment of the present invention the polymerizable substance is a bio-based polymerizable substance. In one embodiment of the present invention the polymerizable substance is selected from a group consisting of lignin and tannin.

In one embodiment of the present invention step (i) comprises tannin as a reactant component.

In one embodiment of the present invention the tannin used originates from any wood species. Tan-

nin may originate from e.g. bark or heartwood. Quebracho tree, beech tree and wattle tree are presented as examples of possible sources of tannin.

In one embodiment of the present invention
5 the tannin used originates from softwood bark. In one embodiment of the present invention the tannin is separated from softwood bark of debarking units in sawmills or pulp mills. The separation process can be combined with an ethanol extraction process, a hot water
10 extraction process, a hot steam extraction process or a water-ethanol extraction process of softwood bark.

In one embodiment of the present invention the tannin is condensed tannin. Condensed tannin has a
15 high dry content and is therefore suitable to be used in the present invention. The dry matter content of condensed tannin may vary between 40 - 100 % and is suitably between 60 - 90 % and preferably between 70 - 80 %. Tannin with such dry matter content can easily
20 be dispersed, whereby a good reactivity with the other reactant components is achieved. The tannin may also be hydrolysable tannin.

In one embodiment of the present invention the method comprises dispersing tannin before adding
25 it to the composition. If the amount of tannin to be used in the production of the binder composition is more than 3 %, preferably more than 5 %, more preferably more than 8 % of the total dry matter content of the components to be used for the production of the
30 binder composition, then tannin is dispersed before it is added to the composition.

The method of the present invention surprisingly results in a more environmentally friendly binder composition since in the method the natural polymer
35 lignin, which is a phenolic polymer, has replaced at least part of the synthetic phenol substance usually used in the production of phenolic compositions such

as phenol formaldehyde resin. Without limiting the invention to any specific theory about why the method of the present inventions results in the aforementioned advantage, it is to be considered that the suitability of replacing at least part of e.g. the phenol with lignin is due to the fact that lignin reacts with an aldehyde, such as formaldehyde, in a quite similar manner as phenol. The use of especially low molecular weight lignin as a reactive additive in the method of the present invention boosts the gluing properties of this kind of lignin based phenolic composition.

In one embodiment of the present invention the catalyst comprises a salt or a hydroxide of an alkali metal. In one embodiment of the present invention the catalyst is selected from a group consisting of sodium hydroxide, potassium hydroxide, acids and their combinations. In one embodiment of the present invention the catalyst is sodium hydroxide.

The precise order of combining and/or adding the components needed for the binder composition production may vary depending e.g. on the required properties of the formed binder composition. The choice of the sequence of combining and/or adding the required components is within the knowledge of the skilled person based on this specification. The precise amount of the components used for producing the binder composition may vary and the choice of the amounts of the different components is within the knowledge of the skilled person based on this specification.

When determining the order of mixing and combining together the components to be used in the production of the binder composition, it should be taken into consideration that low molecular weight lignin is more reactive than high molecular weight lignin. Therefore, high molecular weight lignin may preferably be cooked in the aqueous composition for a longer period of time than the low molecular weight lignin. In

this way it is ensured that the high molecular weight lignin has sufficiently time to react with the cross-linking agent, e.g. the aldehyde.

5 The present invention further relates to a binder composition obtainable by the method according to the present invention.

10 The present invention further relates to an adhesive composition comprising the binder composition according to the present invention. The adhesive composition can further comprise one or more adhesive components selected from a group consisting of other binders, extenders, additives, catalysts and fillers. A binder is a substance, which is mainly responsible for creating the growing and cross-linking of polymer and thus assists in the curing of polymer systems. An extender is a substance, which assists the binder by adjusting physical properties for example by binding moisture. The additive can be a polymer or an inorganic compound, which assists in properties like filling, softening, reducing costs, adjusting moisture, increasing stiffness and increasing flexibility. The catalyst is a substance, which usually boosts and adjusts the curing speed. By "substance" is herein to be understood as including a compound or a composition. 25 The binder composition of the present invention may serve as a binder, an extender, an additive, a catalyst and/or a filler in the adhesive composition.

30 In one embodiment of the present invention a layered composite structure can be formed of two or more layers including at least one wood veneer layer, wherein the layers are arranged the one above the other and combined by means of gluing with the binder composition according to the present invention and/or the adhesive composition according to the present invention. In this specification, unless otherwise stated, the term "wood veneer" is used to address a veneer, which can be formed of any material, e.g. wood-

based material, fiber material, composite material or the like. In this context, the thickness of the wood veneer can be varied. Typically the thickness of wood veneer is below 3 mm.

5 In one embodiment of the present invention the layered composite structure is selected from a group consisting of a wood panel product, a plywood product, a composite product, and a pressed panel product. The layered composite structure can be formed
10 of a number of layers, preferably wood veneer layers, in which the layers are laid one upon the other and glued together.

 The present invention further relates to the use of the binder composition in an impregnation appli-
15 cation, as a coating, for strengthening plastic, for producing a compressed casting, a moulding, a laminate or a lacquer, or for gluing a wood product. The binder composition of the present invention can further be used for gluing combinations of plastic and wood.

20 The present invention further relates to the use of the adhesive composition for gluing a wood product.

 In one embodiment of the present invention the wood product is selected from a group consisting
25 of a wood board, a wood veneer, and a wood bar.

 The embodiments of the invention described hereinbefore may be used in any combination with each other. Several of the embodiments may be combined together to form a further embodiment of the invention.
30 A method, a composition or a use, to which the invention is related, may comprise at least one of the embodiments of the invention described hereinbefore.

 An advantage of the method according to the present invention is that by using both high molecular
35 weight lignin and low molecular weight lignin as reactant components during the production of a binder composition a more environmentally friendly binder compo-

sition is achieved. Surprisingly it has been found out that when using both high molecular weight lignin and low molecular weight lignin as reactant components the amount of the polymerizable substance, such as the synthetic phenol substance, e.g. phenol, can be markedly decreased during the binder production process. As the phenol being a synthetic compound and lignin being a natural polymer, it is advantageous to be able to minimize the amount of phenol present in the final binder composition.

An advantage of the method according to the present invention is that by using low molecular weight lignin in the binder production method the compatibility and reaction behavior of the formed binder composition is better than when using high molecular weight lignin only.

An advantage is of the method according to the present invention is that the use of the more reactive low molecular weight lignin in the binder composition enhances e.g. curing and adhesion properties, and the tensile strength performance of the binder composition. I.e. the overall performance of the formed binder composition is advantageous.

Thus, an advantage of the present invention is that a higher level of bio-based components is achieved in the final binder composition. This advantage can be achieved when using the specific process steps of the present invention and especially when low molecular weight lignin is added to the composition.

EXAMPLES

Reference will now be made in detail to the embodiments of the present invention, an example of which is illustrated in the accompanying drawing.

The description below discloses some embodiments of the invention in such a detail that a person

skilled in the art is able to utilize the invention based on the disclosure. Not all steps of the embodiments are discussed in detail, as many of the steps will be obvious for the person skilled in the art based on this specification.

Figure 1 illustrates a method according to one embodiment of the present invention for producing a binder composition.

Before forming an aqueous composition including the reactant components, the source and amounts of the components are chosen in order to prepare a binder composition with desired properties. Especially the amounts and the source of high molecular weight lignin and low molecular weight lignin are selected.

Following various preparations, step (i) and especially step (ia) of said step is carried out by forming a aqueous dispersion comprising lignin. In the embodiment of Fig. 1 both high molecular weight lignin and low molecular weight lignin are used in step (ia) for forming a lignin dispersion.

A dispersion in step ia) can be formed e.g. by combining water, sodium hydroxide and the lignin fractions under heating at a temperature of e.g. 25 - 60 °C.

Alternatively, if the lignin is to be alkalated in order to make the lignin even more reactive during the binder cooking step, step (ia) can be performed by firstly mixing water and sodium hydroxide and heating the mixture up to a temperature of about 60 °C. Then, high molecular weight lignin can be dispersed into the composition after which low molecular lignin is dispersed. Then the formed dispersion is allowed to cook at a temperature of about 75 °C for about one hour in order to form alkalated lignin. Then, the temperature of the dispersion is decreased to about 50 °C for further use.

Having formed the lignin dispersion in step (ia) the polymerizable substance, the crosslinking agent and the catalyst are added in a predetermined order with simultaneously controlling the temperature of the formed aqueous composition (step (ib)). The selected components can be added one after the other or at least partly simultaneously.

After step (i) of forming the aqueous composition, step (ii) is carried out. Step (ii) comprises cooking the aqueous composition at a temperature of 50 - 95 °C until a predetermined viscosity of the composition is reached.

As a result of step (ia), step (ib), and step (ii) a binder composition having desired properties is produced. This binder composition can be used as such for gluing applications or it can be further processed with other adhesive components for producing an adhesive composition.

Figure 2 illustrates a method according to another embodiment of the present invention for producing a binder composition.

The method according to the embodiment of Fig. 2 begins in a similar manner as in the embodiment of Fig. 1. I.e. either an alkalated lignin dispersion or a non-alkalated lignin dispersion is formed as explained above in relation to Fig 1. However, in the embodiment of Fig. 2 only high molecular weight lignin is used for forming the dispersion in step (ia).

After forming the dispersion in step (ia), step (ib) is carried out. Step (ib) comprises adding the polymerizable substance, the crosslinking agent and the catalyst for forming an aqueous composition. After mixing these components the composition is cooked in step (ic) until the viscosity of the composition is 15 - 70 % of a predetermined viscosity value of the final binder composition. The composition can be cooked at a temperature of 20 - 95 °C.

Then, step (id) of adding low molecular weight lignin to the composition is carried out. The temperature of the formed aqueous composition is increased to 80 - 90 °C and the composition is cooked at this temperature until a predetermined viscosity of the composition is reached or until a desired polymerizing length is achieved (step (ii) of Fig. 2).

As a result of step (ia), step (ib), step (ic), step (id), and step (ii) a binder composition having desired properties is produced. This binder composition can be used as such for gluing applications or it can be further processed with other adhesive components for producing an adhesive composition.

EXAMPLE 1 - Preparing a binder composition

In this example a binder composition was produced. The following components and their amounts were used:

	concentration	amount (g)
water		352
NaOH-I	50 %	181
25 HMW-lignin	98 %	100
LMW-lignin	98 %	200
phenol	90 %	327
formaldehyde	40 %	608
NaOH-II	50 %	90

30 Firstly, water, the first part of NaOH (NaOH-I) and high molecular weight lignin (HMW-lignin) were mixed such that a dispersion was formed. Simultaneously the temperature increased from room temperature to about 60 °C. Then part of the low molecular weight lignin (LMW-lignin) was added to the dispersion while keeping the temperature at 25 - 50 °C.

Then all of the phenol was added followed by adding the formaldehyde in a stepwise manner for forming an aqueous composition. The temperature was kept under 75 °C. After adding the formaldehyde the composition was allowed to react for 10 minutes after which the rest of the low molecular weight lignin was added. After adding also the second part of the NaOH (NaOH-II) to the composition, the composition was cooked at 85 °C until the viscosity of the formed composition was about 305 cp. Then the composition was cooled and the final viscosity was 315 cP. The viscosity was measured at a temperature of 25 °C.

In this example firstly a dispersion of water, NaOH and the lignin was formed after which the addition of phenol was started. However, the dispersion could also have been subjected to an alkalation treatment in order to make the lignin used even more reactive. The alkalation can be performed by firstly mixing water and NaOH, after which heating of the mixture is started. When the temperature has reached 60 °C, the high molecular weight lignin is added followed by the low molecular weight lignin. Then the temperature can be reached up 75 °C and the dispersion is allowed to react for about one hour at this temperature for alkalating the lignin. The dispersion is then cooled down to 50 °C before the addition of phenol is begun.

The procedure of example 1 results in the possibility of replacing 50 % of the synthetic phenol usually used in the binder production method with lignin.

EXAMPLE 2 - Preparing a binder composition

In this example a binder composition was produced. The following components and their amounts were used:

		concentration	amount (g)
	water		578
5	NaOH-I	50 %	254
	HMW-lignin	61 %	492
	LMW-lignin	90 %	111
	phenol	90 %	443
	formaldehyde-I		
10	formaldehyde-II	40 %	448
	NaOH-II	50 %	127

15 Firstly, water and the first part of NaOH (NaOH-I) were mixed and heated up to a temperature of 75 °C. Then the HMW-lignin was added followed by the addition of LMW-lignin. When also the LMW-lignin had been added to the dispersion, the dispersion was heated at a temperature of 75 °C for about one hour in order to form alkalated lignin.

20 After the dispersion had been cooled down to 50 °C the phenol was added followed by adding the first part of the formaldehyde (formaldehyde-I) in a stepwise manner for forming the aqueous composition. Then the temperature was increased up to 75 °C, after which also the other part of the formaldehyde (formaldehyde-II) and the second part of the NaOH (NaOH-II) were added. Then the composition was cooked at 85 °C until the viscosity of the formed composition was about 380 cp. Then the composition was cooled and the final viscosity was 430 cP. The viscosity was measured at a temperature of 25 °C.

30 The procedure of example 2 results in the possibility of replacing 50 % of the synthetic phenol usually used in the binder production method with lignin.

EXAMPLE 3 - Preparing a binder composition

In this example a binder composition was produced. The following components and their amounts were used:

	concentration	amount (g)
10 water		179
NaOH-I	50 %	102
HMW-lignin	97 %	146
LMW-lignin	97%	72
phenol	90 %	123
15 formaldehyde	40 %	370
NaOH-II	50 %	51

Firstly, water, the first part of NaOH (NaOH-I) and HMW-lignin were mixed under heating such that a dispersion was formed. Then the temperature was adjusted to 50 °C, the phenol was added and then the formaldehyde was added in a stepwise manner during a period of one hour. The temperature was kept under 75 °C. After the formaldehyde had been added the second part of the NaOH (NaOH-II) was added. After the addition of NaOH-II, the composition was cooked at 75 °C until the viscosity of the composition was 100 cP. Then the LMW-lignin was added to the composition. The cooking was continued at the temperature of 75 °C until the viscosity of the formed composition was about 305 cP. Then the composition was cooled resulting in a final viscosity of 350 cP. The viscosity was measured at a temperature of 25 °C.

The procedure of example 3 results in the possibility of replacing 66 % of the synthetic phenol usually used in the binder production method with lignin.

EXAMPLE 4 - Preparing a binder composition

In this example a binder composition was produced. The following components and their amounts were used:

	concentration	amount (g)
10 water		137
NaOH-I	50 %	116
HMW-lignin	70 %	91,4
LMW-lignin	90 %	285
phenol	90 %	89
15 formaldehyde	40%	342
NaOH-II	50 %	58

Firstly, water, the first part of NaOH (NaOH-I) and HMW-lignin were mixed such that a dispersion was formed. Simultaneously the temperature increased from room temperature to about 60 °C. Then the LMW-lignin was added to the dispersion while keeping the temperature at 25 - 50 °C.

Then all of the phenol was added followed by adding the formaldehyde in a stepwise manner for forming an aqueous composition. The temperature was kept under 75 °C. After adding the formaldehyde the composition was allowed to react for 10 minutes after which also the second part of the NaOH (NaOH-II) to the composition. Then the composition was cooked at 70 - 80 °C until the viscosity of the formed composition was about 375 cp. Then the composition was cooled and the final viscosity was 390 cP. The viscosity was measured at a temperature of 25 °C.

In this example firstly a dispersion of water, NaOH and both lignin fractions were formed after which the addition of phenol was started. However, the

dispersion could also have been subjected to alkala-
tion treatment in accordance with the procedure pre-
sented in example 1 in order to make the lignin used
even more reactive.

5 The procedure of example 4 results in the
possibility of replacing 80 % of the synthetic phenol
usually used in the binder production method with lig-
nin.

10 EXAMPLE 5 - Preparing an adhesive composition

 In this example the binder composition pro-
duced in Example 1 was used for the production of an
adhesive composition. The binder composition was mixed
15 with extenders, fillers, catalysts, additives, as ex-
amples of which e.g. starch, wood flour and hardener
(e.g. tannin or carbonates) can be mentioned, thus
forming the adhesive composition.

20 EXAMPLE 6 - Applying the binder composition for pro-
ducing a plywood product

 Wood veneers having the thickness of below 3
mm were glued together with the binder composition
25 produced in Example 2 for producing a 7-plywood. Re-
sults showed that the gluing effect was sufficiently
good for gluing wood veneers.

30 EXAMPLE 7 - Applying the adhesive composition for pro-
ducing a plywood product

 In this example the adhesive composition of
Example 5 was applied onto wood veneers. The wood ve-
neers were joined together by the adhesive composition
35 for forming a plywood. The dry matter content of the
adhesive composition was between 45 and 55 %. The wood
veneers with the adhesive composition were pressed by

hot-pressing technique at a temperature between 120 -
170 °C. The adhesive composition was simultaneously
cured. The adhesive composition of the present inven-
tion was found suitable for gluing wood veneers to-
5 together and thus for manufacturing plywood.

EXAMPLE 8 - Applying the binder composition for pro-
ducing laminates

10 In this example the binder composition as
produced in Example 3 was used in an impregnation ap-
plication. During the production of laminates paper
was impregnated with an alcohol solution of the binder
composition, after which the impregnated layers were
15 transferred into a furnace. The alcohol was volati-
lized and the binder composition was partly cured. The
layers comprising such semi-cured composition were ar-
ranged the one above the other and baked by a hot-
pressing technique in order to form uniform thicker
20 boards or laminates.

In the binder production method presented in
the examples above, phenol and formaldehyde are used
are used as examples of polymerizable substance and
25 crosslinking agent, respectively. However, any other
polymerizable substance or crosslinking agent dis-
closed in this specification can be equally well used
in the binder composition production method as will be
obvious for the skilled person based on this specifi-
30 cation.

It is obvious to a person skilled in the art
that with the advancement of technology, the basic
idea of the invention may be implemented in various
ways. The invention and its embodiments are thus not
35 limited to the examples described above; instead they
may vary within the scope of the claims.

CLAIMS

1. A method for producing a binder composition, characterized in that the method comprises the following steps:

5 (i) forming an aqueous composition comprising reactant components including lignin molecules of 11 - 60 lignin units, lignin molecules of 1 - 10 lignin units, polymerizable substance and crosslinking agent in the presence of a catalyst, wherein the polymerizable substance is selected from a group consisting of phenol, cresol, resorcinol and combinations thereof, and the crosslinking agent is an aldehyde; and

10 (ii) cooking the composition at a temperature of 45 - 95 °C for polymerizing the reactant components until a binder composition with a predetermined viscosity value is formed.

2. The method of claim 1, characterized in that the weight ratio of lignin molecules of 11 - 60 lignin units to lignin molecules of 1 - 10 lignin units is 0,1 - 20, preferably 0,2 - 10, and more preferably 0,5 - 5.

3. The method of any one of claims 1 - 2, characterized in that step (i) of forming the aqueous composition comprises the following steps:

25 (ia) forming a dispersion comprising lignin molecules of 11 - 60 lignin units; and

(ib) combining polymerizable substance and crosslinking agent with the dispersion for forming the aqueous composition.

30 4. The method of claim 3, characterized in that step (ib) comprises adding catalyst.

5. The method of any one of claims 3 - 4, characterized in that step (i) of forming the aqueous composition comprises, after step (ib), the following steps:

35 (ic) cooking the formed composition until the composition has a viscosity, which corresponds to 15 -

70 % of a predetermined viscosity value of the final binder composition; and

(id) adding lignin molecules of 1 - 10 lignin units as a reactant component to the composition.

5 6. The method of any one of claims 3 - 5, characterized in that step (ia) and/or step (ib) comprises adding lignin molecules of 1 - 10 lignin units as a reactant component.

10 7. The method of any one of claims 1 - 6, characterized in that step (i) comprises keeping the composition at a temperature of at most 75 °C.

15 8. The method of any one of claims 5 - 7, characterized in that step (ic) comprises keeping the temperature of the composition between 45 - 95 °C, preferably between 50 - 85 °C, and more preferably between 60 - 80 °C.

20 9. The method of any one of claims 5 - 8, characterized in that the temperature of the composition is decreased below 85 °C, and preferably below 65 °C before step (id).

25 10. The method of any one of claims 1 - 9, characterized in that step (ii) comprises cooking the composition preferably at a temperature of 50 - 85 °C, and more preferably at a temperature of 60 - 80 °C.

30 11. The method of any one of claims 5 - 10, characterized in that the temperature of the composition is increased up to 45 - 95 °C, preferably up to 50 - 85 °C, and more preferably up to 60 - 80 °C, before, during or after step (id).

35 12. The method of any one of claims 1 - 11, characterized in that the weight ratio of lignin molecules of 11 - 60 lignin units and lignin molecules of 1 - 10 lignin units to the polymerizable substance is at least 0,2, preferably at least 1, and more preferably at least 5.

13. The method of any one of claims 1 - 12,
c h a r a c t e r i z e d in that the crosslinking agent
is preferably formaldehyde.

14. The method of any one of claims 1 - 13,
5 c h a r a c t e r i z e d in that the catalyst is sodium
hydroxide.

PATENTTIVAATIMUKSET

1. Menetelmä sideainekoostumuksen valmistamiseksi, tunnettu siitä, että menetelmä käsittää seuraavat vaiheet:

- 5 (i) muodostetaan vesipitoinen koostumus, joka käsittää reagenssikomponentteja, jotka sisältävät 11 - 60 ligniiniyksikön ligniinimolekyylejä, 1 - 10 ligniiniyksikön ligniinimolekyylejä, polymerisoituvaa ainetta ja silloittavaa ainetta, katalyytin läsnä ollessa, 10 jossa polymerisoituva aine on valittu joukosta, joka koostuu fenolista, kresolista, resorsinolista ja niiden yhdistelmistä, ja silloittava aine on aldehydi; ja
- (ii) keitetään koostumusta 45 - 95 °C:n lämpötilassa reagenssikomponenttien polymerisoimiseksi, 15 kunnes muodostuu sideainekoostumus, jolla on ennalta määrätty viskositeettiarvo.

2. Patenttivaatimuksen 1 mukainen menetelmä, tunnettu siitä, että 11 - 60 ligniiniyksikön ligniinimolekyyleiden painosuhte 1 - 10 ligniiniyksikön 20 ligniinimolekyyleihin on 0,1 - 20, edullisesti 0,2 - 10 ja edullisemmin 0,5 - 5.

3. Jonkin patenttivaatimuksista 1 - 2 mukainen menetelmä, tunnettu siitä, että vaihe (i), jossa muodostetaan vesipitoinen koostumus, käsittää 25 seuraavat vaiheet:

- (ia) muodostetaan dispersio, joka käsittää 11 - 60 ligniiniyksikön ligniinimolekyylejä; ja
- (ib) lisätään dispersioon polymerisoituvaa ainetta ja silloittavaa ainetta vesipitoisen koostumuksen muodostamiseksi. 30

4. Patenttivaatimuksen 3 mukainen menetelmä, tunnettu siitä, että vaihe (ib) käsittää katalyytin lisäämisen.

5. Jonkin patenttivaatimuksista 3 - 4 mukainen 35 menetelmä, tunnettu siitä, että vaihe (i),

jossa muodostetaan vesipitoinen koostumus, käsittää vaiheen (ib) jälkeen seuraavat vaiheet:

(ic) keitetään muodostunutta koostumusta, kunnes koostumuksen viskositeetti vastaa 15 - 70 %:a
5 lopullisen sideainekoostumuksen ennalta määrätystä viskositeetti-arvosta; ja

(id) lisätään koostumukseen 1 - 10 ligniiniyksikön ligniinimolekyylejä reagenssikomponenttina.

6. Jonkin patenttivaatimuksista 3 - 5 mukainen menetelmä, tunnettu siitä, että vaihe (ia)
10 ja/tai vaihe (ib) käsittää 1 - 10 ligniiniyksikön ligniinimolekyylien lisäämisen reagenssikomponenttina.

7. Jonkin patenttivaatimuksista 1 - 6 mukainen menetelmä, tunnettu siitä, että vaihe (i) kä-
15 sittää koostumuksen pitämisen korkeintaan 75 °C:n lämpötilassa.

8. Jonkin patenttivaatimuksista 5 - 7 mukainen menetelmä, tunnettu siitä, että vaihe (ic)
20 käsittää koostumuksen lämpötilan pitämisen 45 - 95 °C:n välillä, edullisesti 50 - 85 °C:n välillä ja edullisemmin 60 - 80 °C:n välillä.

9. Jonkin patenttivaatimuksista 5 - 8 mukainen menetelmä, tunnettu siitä, että koostumuksen
25 lämpötila lasketaan alle 85 °C:een ja edullisesti alle 65 °C:een ennen vaihetta (id).

10. Jonkin patenttivaatimuksista 1 - 9 mukainen menetelmä, tunnettu siitä, että vaihe (ii)
30 käsittää koostumuksen keittämisen edullisesti 50 - 85 °C:n lämpötilassa ja edullisemmin 60 - 80 °C:n lämpötilassa.

11. Jonkin patenttivaatimuksista 5 - 10 mukainen menetelmä, tunnettu siitä, että koostumuksen
35 lämpötila nostetaan 45 - 95 °C:een, edullisesti 50 - 85 °C:een ja edullisemmin 60 - 80 °C:een ennen vaihetta (id), sen aikana tai sen jälkeen.

12. Jonkin patenttivaatimuksista 1 - 11 mukainen menetelmä, tunnettu siitä, että 11 - 60 ligniiniyksikön ligniinimolekyylien ja 1 - 10 ligniiniyksikön ligniinimolekyylien painosuhde polymerisoi-
5 tuvaan aineeseen on vähintään 0,2, edullisesti vähintään 1 ja edullisemmin vähintään 5.

13. Jonkin patenttivaatimuksista 1 - 12 mukainen menetelmä, tunnettu siitä, että silloittava aine on edullisesti formaldehydi.

10 14. Jonkin patenttivaatimuksista 1 - 13 mukainen menetelmä, tunnettu siitä, että katalyytti on natriumhydroksidi.

15

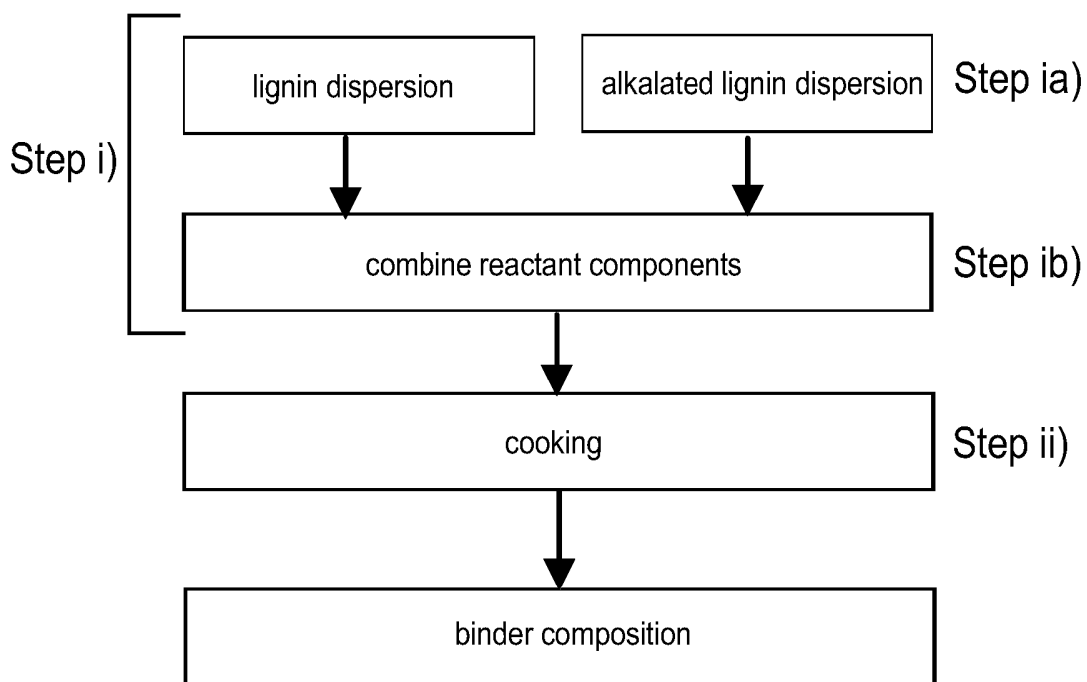


Fig. 1

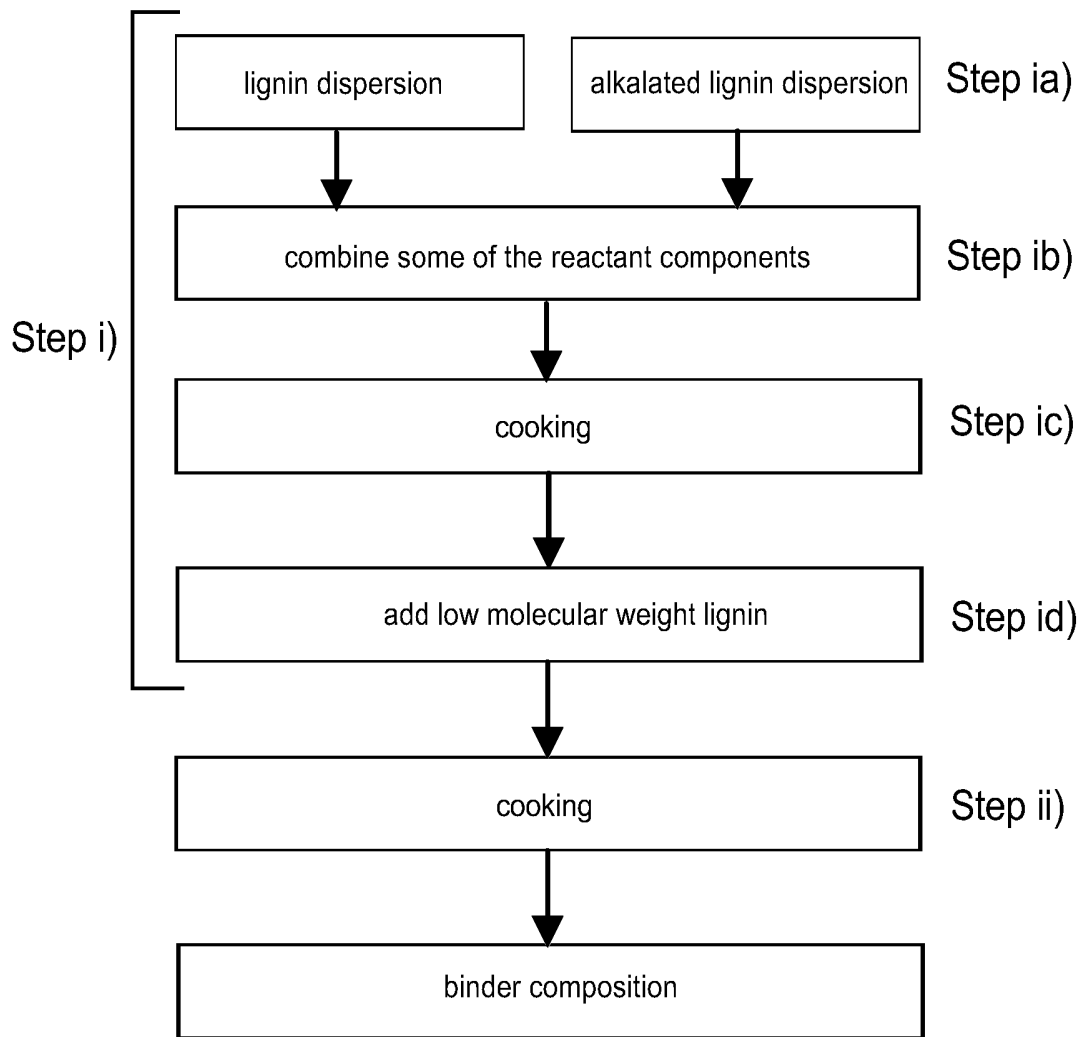


Fig. 2