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(72) Inventeurs/Inventors:
PIETARINEN, SUVI, FI;
VALKONEN, SANNA, FI;
RINGENA, OKKO, DE
(73) Propriétaire/Owner:
UPM-KYMMENE CORPORATION, FI
(74) Agent: SIM & MCBURNEY

(54) Titre : PROCEDE DE TRAITEMENT DE LIGNINE ET DE PRODUCTION D'UNE COMPOSITION LIANTE
(54) Title: A METHOD FOR TREATING LIGNIN AND FOR PRODUCING A BINDER COMPOSITION

(57) **Abrégé/Abstract:**

The present invention relates to a method for treating lignin, wherein the method comprises the following steps: a) dissolving lignin into an aqueous composition, which contains a compound selected from the class of phenols and alkali, while keeping the temperature of the composition at 40 - 85 °C, wherein the alkali comprises a hydroxide of an alkali metal; and b) heating the composition at a temperature, which is higher than the temperature of the composition in step a), with the proviso that the temperature of the composition does not exceed 100 °C, while keeping the pH of the composition at a pH value of 6 - 14.

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(71) **Applicant:** UPM-KYMMENE CORPORATION
[FI/FI]; Alvar Aallon katu 1, FI-00100 Helsinki (FI).

(72) **Inventors:** PIETARINEN, Suvi; Mäkikatu 21 A, FI-53100 Lappeenranta (FI). VALKONEN, Sanna; Tuulimyllynkatu 46 B 9, FI-53500 Lappeenranta (FI). RINGENA, Okko; Ganghoferstrasse 3, 86356 Neusäss (DE).

(74) **Agent:** PAPULA OY; P.O. Box 981, FI-00101 Helsinki (FI).

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(54) **Title:** A METHOD FOR TREATING LIGNIN AND FOR PRODUCING A BINDER COMPOSITION

(57) **Abstract:** The present invention relates to a method for treating lignin, wherein the method comprises the following steps: a) dissolving lignin into an aqueous composition, which contains a compound selected from the class of phenols and alkali, while keeping the temperature of the composition at 40 - 85 °C, wherein the alkali comprises a hydroxide of an alkali metal; and b) heating the composition at a temperature, which is higher than the temperature of the composition in step a), with the proviso that the temperature of the composition does not exceed 100 °C, while keeping the pH of the composition at a pH value of 6 - 14.

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A METHOD FOR TREATING LIGNIN AND FOR PRODUCING A BINDER COMPOSITION

FIELD OF THE INVENTION

5 The invention relates to a method for treating lignin and to the use thereof for producing a binder composition as well as to different applications thereof.

BACKGROUND OF THE INVENTION

 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
15 come up with a more environmentally friendly adhesive composition. Especially, the ability to replace synthetic phenol in phenolic resins, such as phenol formaldehyde resin, has been the object of prior art. Lignin can be used for the purpose of decreasing the
20 amount of synthetic phenol in a resin composition. Lignin has previously been used for replacing phenol during the production of lignin-phenol-formaldehyde resin.

 The currently known lignin based resins are
25 not, however, suitable for all the applications where traditional phenolic resins are being used. For example, the currently known lignin based resins are not suitable for high-pressure laminates. High-pressure laminates (HPL), which are also known as plastic laminates, can be produced by fusing together under the
30 influence of heat and pressure, multiple layers of paper, fabrics, or other core materials by using thermosetting resins as binders. The inventors have recognized a need for a method, which would result in a
35 higher phenol replacement in the resin and thus in a more environmentally friendly binder composition hav-

ing suitable properties for use in different applications such as high-pressure laminates.

PURPOSE OF THE INVENTION

5 The purpose of the invention is to provide a new type of method for treating lignin and to provide a method for producing a binder composition for different applications. The purpose of the invention is to provide a new binder composition and adhesive composition as well as the uses thereof.
10

SUMMARY

 Accordingly, in one aspect there is provided a method for treating lignin, comprising: a) dissolving
15 ing lignin into an aqueous composition, which contains a compound selected from a class of phenols and alkali, while keeping a temperature of the composition between 40°C and 85°C and the pH of the composition at a pH value of between 4 and 10, wherein the alkali comprises a hydroxide of an alkali metal and wherein the
20 compound selected from the class of phenols is selected from a group consisting of phenol, cresol, resorcinol and combinations thereof; and b) heating the composition at a temperature, which is higher than the
25 temperature of the composition in step a), with the proviso that the temperature of the composition does not exceed 100°C, while keeping the pH of the composition at a pH value of between 6 and 10, for allowing lignin to react with the compound selected from the
30 class of phenols and the alkali.

According to another aspect there is provided a method for producing a binder composition, comprising: a) dissolving lignin into an aqueous composition, which contains a compound selected from a class of phenols and alkali, while keeping a temperature of the composition at between 40°C and 85°C and a pH of the composition at a pH value of between 4 and 10, wherein the alkali comprises a hydroxide of an alkali metal and wherein the compound selected from the class of phenols is selected from a group consisting of phenol, cresol, resorcinol and combinations thereof; b) heating the composition at a temperature, which is higher than the temperature of the composition in step a), with the proviso that the temperature of the composition does not exceed 100°C, while keeping the pH of the composition at the pH value between 6 and 10, for allowing lignin to react with the compound selected from the class of phenols and the alkali; and c) mixing the composition formed in step b) with a cross-linking agent and heating the composition at a temperature of between 60°C and 100°C while keeping the pH of the composition at the pH value between 6 and 10.

BRIEF DESCRIPTION OF THE DRAWINGS

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

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

Fig. 2 is a flow chart illustration of a method for producing a binder composition according to one embodiment of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

The present invention relates to a method for treating lignin, which method comprises the following steps:

a) dissolving lignin into an aqueous composition, which contains a compound selected from the class of phenols and alkali, while keeping the temperature of the composition at 40 - 85 °C, wherein the alkali comprises a hydroxide of an alkali metal; and

b) heating the composition at a temperature, which is higher than the temperature of the composition in step a), with the proviso that the temperature of the composition does not exceed 100 °C, while keeping the pH of the composition at a pH value of 6 - 14.

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

a) dissolving lignin into an aqueous composition, which contains a compound selected from the class of phenols and alkali, while keeping the temperature of the composition at 40 - 85 °C, wherein the alkali comprises a hydroxide of an alkali metal;

b) heating the composition at a temperature, which is higher than the temperature of the composition in step a), with the proviso that the temperature of the composition does not exceed 100 °C, while keeping the pH of the composition at a pH value of 6 - 14; and

c) mixing the composition formed in step b) with a crosslinking agent and heating the composition

at a temperature of 60 - 100 °C while keeping the pH of the composition at a pH value of 6 - 14.

In one embodiment of the present invention the compound selected from the class of phenols is selected from a group consisting of phenol, cresol, resorcinol and combinations thereof. In one embodiment of the present invention the compound selected from the class of phenols is phenol.

In one embodiment of the present invention lignin is dissolved into a composition, which consists of water, at least one compound selected from the class of phenols, and alkali, in step a). In one embodiment of the present invention lignin is dissolved into an aqueous composition of a compound selected from the class of phenols and alkali.

In one embodiment of the present invention the temperature of the composition is kept, in step a), at a temperature of 65 - 80, and preferably at a temperature of 70 - 75 °C.

In one embodiment of the present invention step b) comprises heating the composition at a temperature, which is at least 5 °C, preferably at least 10 °C, and more preferably at least 20 °C higher than the temperature of the composition in step a).

In one embodiment of the present invention the composition is heated, in step b), at a temperature of 60 - 100 °C, preferably at a temperature of 70 - 95 °C, and more preferably at a temperature of 75 - 90 °C.

In one embodiment of the present invention step b) is carried out for allowing lignin to react with the compound selected from the class of phenols and the alkali.

In one embodiment of the present invention further alkali is added to the composition in step b). In one embodiment of the present invention alkali is added to the composition in step a) and in step b).

The inventors of the present invention surprisingly found out that dissolving lignin into a composition of e.g. phenol, alkali and water, at the specific temperature used, advantageously affects the time needed to dissolve lignin into the composition. The inventors of the present invention found out that treating lignin in accordance with step a) and step b) enables the production of a binder composition with a low pH value if desired. The inventors of the present invention found out that by the method of the present invention it was possible to produce a lignin based binder composition that is suitable for e.g. the production of structural wood panel products, and impregnated overlay materials. The inventors also found out that different types of lignin, such as dry or condensed lignin that are usually challenging and time-consuming to dissolve, could be more easily dissolved by the method according to the present invention. Also, the inventors noticed that the alkali used for activating lignin was not consumed during the step of dissolving lignin, whereby less chemical is needed thus lower the overall chemical costs.

In one embodiment of the present invention step b) is carried out for alkalating the lignin. The alkalation step results in the lignin being activated. Without limiting the invention to any specific theory about why alkalation of lignin results in a more activated or more reactive lignin being formed compared to non-alkalated lignin, it is to be considered that the alkalation opens the macromolecular structure of lignin whereby the steric hindrances that usually disable reactive groups in lignin structures are removed. Alkalation may also add charged groups to the lignin macromolecule. In one embodiment of the present invention treating lignin with the method according to the present invention activates the lignin. The advantage of using alkalated lignin e.g. for producing a binder

composition is that the compatibility and reaction behavior is much better than in a normal case, where non-treated lignin has been used in the cooking or polymerizing stage of production process.

5 In this specification, unless otherwise stated, the expression "lignin" should be understood as lignin originating from any suitable lignin source.

The lignin used can be essentially pure lignin. By the expression "essentially pure lignin" 10 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 lignin comprises at most 10 %, preferably at most 5 %, of other components. Extractives and carbohydrates 15 such as hemicelluloses can be mentioned as examples of such other components. In one embodiment of the present invention the lignin contains less than 10 weight-%, preferably less than 6 weight-%, and more preferably less than 4 weight-% of carbohydrates. The 20 amount of carbohydrates present in lignin can be measured by high performance anion exchange chromatography with pulsed amperometric detector (HPAE-PAD) in accordance with standard SCAN-CM 71.

In one embodiment of the present invention 25 the ash percentage of lignin is less than 7.5 weight-%, preferably less than 5 weight-%, and more preferably less than 3 weight-%. The ash content can be determined by carbonifying and quickly burning a lignin sample so that alkali salts are not melted before the 30 organic matter has been burned (e.g. 20 - 200 °C for 30 minutes, after which temperature is adjusted to 200 - 600 °C for 1 h, and thereafter adjusting the temperature to 600 - 700 °C for 1 hour), and finally the lignin sample is ignited at 700 °C for 1h. Ash content 35 of a lignin sample refers to the mass that remains of the sample after burning and ignition, and it is presented as per cent of the sample's dry content.

In one embodiment of the present invention the weight average molecular weight (Mw) of lignin is 1000 - 15000 g/mol, preferably 2000 - 10000 g/mol, and more preferably 3000 - 8000 g/mol. The molecular weight of the lignin can be determined by using a high-performance size-exclusion chromatography in the following manner: Two parallel measurements are carried out. 0.1 M NaOH is used as an eluent. Calibration is done using Na-polystyrene sulfonate standards having a molecular weight of 1100 - 73900 g/mol. For quality control, standard quality kraft lignin and PSS molecular weight standard are used. The columns used are PSS MCX™ precolumns, 1000 Å and 100 000 Å separation columns filled with sulfonated styrene-divinylbenzene copolymer matrix. Isocratic run program is used. The run time is 45 minutes. The injection volume is 50 µl. The flux is 0.5 ml per minute. The temperature is 25 °C. As a result of the chromatography, number average molecular weight (Mn), weight average molecular weight (Mw), peak molecular weight (Mp) and polydispersity index (PDI) values can be reported.

In one embodiment of the present invention the number average molecular weight (Mn) of lignin is 700 - 4000, preferably 800 - 3000, and more preferably 1000 - 2500.

In one embodiment of the present invention the polydispersity of lignin is 1.0 - 7, preferably 1.2 - 6, and more preferably 1.4 - 4.5.

In one embodiment of the present invention the normalized radical scavenger index (NRSI) of lignin is 0.01 - 20, preferably 0.05 - 10, and more preferably 0.1-6. The antioxidant activity of extracts can be evaluated using DPPH-method in methanol extracts. The basis of the radical scavenging method is described in Malterud et al. (Pharmacol. Toxicol. 1996, 78: 111-116). The method is based on the ability of

extracts and pure components to react with 1,1,-
diphenylpicryl-2-hydrazylradical (DPPH•)-molecules
when DPPH loses its radical characteristic. The de-
cline of radical form can be observed with spectropho-
5 tometer as a change of solution color from violet to
yellow (absorbance is measured at wavelength 515 nm).
RSI (radical scavenging index) is defined as the in-
verse of the concentration that produced 50 % inhibi-
tion in DPPH absorbance at 515 nm. The results can
10 then be "normalized" (NRSI) by dividing the sample RSI
by the RSI value for the butyl hydroxytoluene (BHT)
control.

In one embodiment of the present invention
the dry solids content of lignin is below 98 %, pref-
15 erably 40 - 80 %, and more preferably 50 - 70 %. The
dry solids content can be measured by drying a lignin
sample of 1 - 5 g at a temperature of 60 °C or above
in a vacuum oven for four hours.

In one embodiment of the present invention
20 the lignin has an amount of 0.1 - 6 mmol, preferably
0.3 - 3.5 mmol of aliphatic hydroxyl groups per gram
of dry lignin.

In one embodiment of the present invention
the lignin has an amount of 0.1 - 5 mmol, preferably
25 1.5 - 4.5 mmol of phenolic hydroxyl groups per gram of
dry lignin. Aliphatic and phenolic hydroxyl groups can
be determined by characterizing a lignin sample with
31P NMR spectroscopy after phosphitylation and after
which the aliphatic hydroxyl groups can be quantita-
30 tively determined. For 31P NMR analyses 40 mg of lig-
nin can be weighted and dissolved in 300 µl of N,N-
dimethylformamide. After total dissolution 200 µl of
pyridine, 400 µl (0.05M) of internal standard solution
(ISTD) of Endo-N-Hydroxy-5-norbornene-2,3-
35 dicarboximide in pyridine/CDCl₃ and 100 µl of
Cr(acac)₃ solution in pyridine/CDCl₃ is added. Then 200
µl of phosphitylation reagent 2-chloro-4,4,5,5-

tetramethyl-1,3,2-dioxaphospholane is added drop-wise. Finally 600 µl of CDCl₃ is added to the solution and clear brown to black solution is achieved. Freshly prepared samples can then be measured with ³¹P NMR at
5 room temperature. Bruker 500 MHz NMR spectrometer can be used for the measurement. ³¹P NMR measurement is based on the method developed by Grannata and Argyropoulos (Grannata A. and Argyropoulos D.S., 2-Chloro-4,4,5,5-tetramethyl-1,3,2-dioxaphospholane, a reagent
10 for the accurate determination of the uncondensed and condensed phenolic moieties in lignins. J. Agric. Food Chem, 1995, 43:1538-1544). The results are calculated as mmol/g lignin.

In one embodiment of the present invention
15 the lignin is selected from a group consisting of kraft lignin, steam explosion lignin, biorefinery lignin, supercritical separation lignin, hydrolysis lignin, flash precipitated lignin, biomass originating lignin, lignin from alkaline pulping process, lignin
20 from soda process, lignin from organosolv pulping and combinations thereof. In one embodiment of the present invention the lignin is wood based lignin. The lignin can originate from softwood, hardwood, annual plants or from a combination thereof.

25 Different lignin components may have different properties, e.g. molecular weight, molar mass, polydispersity, hemicellulose and extractive contents and compositions.

By "kraft lignin" is to be understood in this
30 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
35 comprises components originating from different softwood and hardwood species in various proportions. Lignin can be separated from the black liquor by differ-

ent techniques including e.g. precipitation and filtration. Lignin usually begins precipitating at pH 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 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 molecular weight distribution of lignin fraction precipitated at a lower pH value is wider than of lignin fraction precipitated at a higher pH value.

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

In one embodiment of the present invention the lignin is flash precipitated lignin. The term "flash precipitated lignin" should be understood in this specification as lignin that has been precipitated from black liquor in a continuous process by decreasing the pH of a black liquor flow, under the influence of an over pressure of 200 - 1000 kPa, down to the precipitation level of lignin using a carbon dioxide based acidifying agent, preferably carbon dioxide, and by suddenly releasing the pressure for precipitating lignin. The method for producing flash precipitated lignin is disclosed in patent application FI 20106073. The residence time in the above method is under 300 s. The flash precipitated lignin particles, having a particle diameter of less than 2 μm , form agglomerates, which can be separated from black liquor using e.g. filtration. The advantage of the flash precipitated lignin is its higher reactivity compared to normal kraft lignin. The flash precipitated lignin can

be purified and/or activated if needed for the further processing.

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

In one embodiment of the present invention the lignin is steam explosion lignin. Steam explosion is a pulping and extraction technique that can be applied to wood and other fibrous organic material.

By "biorefinery lignin" is to be understood in this specification, unless otherwise stated, lignin that can be recovered from a refining facility or process where biomass is converted into fuel, chemicals and other materials.

By "supercritical separation lignin" is to be understood in this specification, unless otherwise stated, lignin that can be recovered from biomass using supercritical fluid separation or extraction technique. Supercritical conditions correspond to the temperature and pressure above the critical point for a given substance. In supercritical conditions, distinct liquid and gas phases do not exist. Supercritical water or liquid extraction is a method of decomposing and converting biomass into cellulosic sugar by employing water or liquid under supercritical conditions. The water or liquid, acting as a solvent, extracts sugars from cellulose plant matter and lignin remains as a solid particle.

In one embodiment of the present invention the lignin is hydrolysis lignin. Hydrolysed lignin can be recovered from paper-pulp or wood-chemical processes.

5 In one embodiment of the present invention the lignin originates from an organosolv process. Organosolv is a pulping technique that uses an organic solvent to solubilize lignin and hemicellulose.

10 The pH value of the composition in step a) and in step b) can be selected depending on the final application of the binder composition to be produced.

In one embodiment of the present invention, the pH of the composition is kept, in step a), at the pH value of 4 - 10, preferably at the pH value of 4.5 - 9.5, more preferably at the pH value of 5 - 9, and even more preferably at the pH value of 5.5 - 8.5. In one embodiment of the present invention the pH of the composition is kept, in step b), preferably at the pH value of 6 - 10, more preferably at the pH value of 6.5 - 9.5, and even more preferably at the pH value of 7 - 9. These kinds of pH ranges may be used in step a) and in step b) when producing a binder composition for high-pressure laminates.

25 In one embodiment of the present invention, the pH of the composition is kept, in step a), at the pH value of 4 - 14, preferably at the pH value of 7 - 13.5, and more preferably at the pH value of 8.5 - 13. In one embodiment of the present invention the pH of the composition is kept, in step b), preferably at the pH value of 7 - 14, more preferably at the pH value of 9 - 13.5, and even more preferably at the pH value of 10 - 13. These kinds of pH ranges may be used in step a) and in step b) when producing a binder composition for plywood.

35 In one embodiment of the present invention step b) is carried out for 10 minutes - 2 hours, preferably for 1 hour - 1.5 hours.

In one embodiment of the present invention the alkali is sodium hydroxide, potassium hydroxide, or a combination thereof.

In one embodiment of the present invention
5 the concentration of alkali is 0.1 - 11 weight-%, and preferably 0.5 - 9 weight-% based on the total weight of the composition in step b). In one embodiment of the present invention the concentration of alkali is 0.1 - 5 weight-%, and preferably 0.5 - 2 weight-%
10 based on the total weight of the composition in step b). In one embodiment of the present invention the concentration of alkali is 3 - 15weight-%, preferably 5 - 12, and more preferably 6 - 10 weight-% based on the total weight of the composition in step b).

15 In one embodiment of the present invention the concentration of lignin in step a) is 10 - 40 weight-%, and preferably 20 - 30 weight-% based on the total weight of the composition in step a).

In one embodiment of the present invention
20 the concentration of the compound selected from the class of phenols in step a) is 10 - 50 weight-%, preferably 20 - 50 weight-%, and more preferably 20 - 45 weight-% based on the total weight of the composition in step a).

25 The step of heating the formed composition in step c) is carried out for polymerizing the reactant components, i.e. lignin, the compound selected from the class of phenols, and the crosslinking agent, such that the viscosity of the binder composition is in-
30 creased. The heating can be continued until a predetermined viscosity value is formed. The predetermined viscosity value of the final binder composition may vary depending on the specific application where the binder composition is to be used.

35 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 embodiment 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 5 50 cP but not more than 150 cP, and more preferably at least 80 but not more than 120 cP.

In one embodiment of the present invention the predetermined viscosity value of the final binder composition is at least 250 cP, preferably at least 10 300 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. 15

The viscosity is measured at 25 °C using a rotary viscometer.

In one embodiment of the present invention, step c) comprises heating the composition preferably 20 at a temperature of 65 - 95 °C, more preferably at a temperature of 70 - 90 °C, and even more preferably at a temperature of 75 - 85 °C.

In one embodiment of the present invention the crosslinking agent is selected from a group consisting of an aldehyde, a derivative of an aldehyde, 25 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 30 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 35 of the present invention the crosslinking agent is an aldehyde, and preferably formaldehyde, paraformaldehyde or a combination thereof.

In one embodiment of the present invention step c) is carried out in the presence of a catalyst. In one embodiment of the present invention the catalyst is selected from a group consisting of sodium hydroxide, potassium hydroxide and any mixture thereof.

In one embodiment of the present invention the pH of the composition is kept, in step c), preferably at a pH value of 6 - 10, more preferably at the pH value of 6.5 - 9.5, and even more preferably at the pH value of 7 - 9.

In one embodiment of the present invention the pH of the composition is kept, in step c), preferably at the pH value of 7 - 14, more preferably at the pH value of 9 - 13.5, and even more preferably at the pH value of 10 - 13.

The present invention further relates to the use of lignin treated by the method according to the present invention for the production of a binder composition.

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

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.

The present invention further relates to the use of a binder composition obtainable by the method according to the present invention, wherein the pH of the composition is kept in step a) at a value of 4 - 10 and in step b) at a value of 6 - 10, for producing a laminate, and preferably a high-pressure laminate.

The present invention further relates to the use of a binder composition obtainable by the method according to the present invention, wherein the pH of

the composition is kept in step c) at a value of 6 - 10 for producing a laminate, and preferably a high-pressure laminate.

5 The present invention further relates to the use of a binder composition obtainable by the method according to the present invention, wherein the pH of the composition is kept in step a) at a value of 4 - 10 and in step b) at a value of 6 - 10, for producing a foundry material, a protective coating, a friction
10 material, an abrasive material, glass wool, rock wool, a prepreg, a shuttering film, an overlay, a molding component or a fiber reinforced composite.

The present invention further relates to the use of a binder composition obtainable by the method
15 according to the present invention, wherein the pH of the composition is kept in step c) at a value of 6 - 10, for producing a foundry material, a protective coating, a friction material, an abrasive material, glass wool, rock wool, a prepreg, a shuttering film,
20 an overlay, a molding component or a fiber reinforced composite.

The present invention further relates to the use of a binder composition obtainable by the method according to the present invention, wherein the pH of
25 the composition is kept in step a) at a value of 4 - 14 and in step b) at a value of 7 - 14, for producing a particle board, an oriented strand board, a chip board, an intrallam, a gluelam, a hardboard, a waferboard, a fiber board, a, plywood, or a wood adhesive.

30 The present invention further relates to the use of a binder composition obtainable by the method according to the present invention, wherein the pH of the composition is kept, in step c), preferably at the pH value of 7 - 14, for producing a particle board, an
35 oriented strand board, a chip board, an intrallam, a gluelam, a hardboard, a waferboard, a fiber board, a, plywood, or a wood adhesive.

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.

5 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 the time needed for dissolving
10 ing lignin into the aqueous composition is reduced when using the specified temperature.

An advantage of the method according to the present invention is that it is possible to treat lignin in a manner that enables the production of a low
15 pH binder composition, i.e. a binder composition with a neutral or acidic pH range. The lignin treated with the method according to the present invention has an open structure making it more reactive whereby the production of low pH binder composition is possible
20 with lignin.

An advantage of the present invention is that a bio-based binder composition suitable to be used for the production of high-pressure laminates can be produced by the method according to the present invention.
25 tion. An advantage of the present invention is that a more environmentally friendly binder composition can be produced for high-pressure laminates.

EXAMPLES

30 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
35 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 treating
5 lignin.

Before the treatment of lignin is started, the source of lignin is chosen. Also the other components and their amounts to be used in the method according to the present invention are selected. If
10 needed, the components used in the method of Fig. 1 can be pre-treated to be suitable for the lignin treatment processes.

Following the various preparations and pre-treatments, in one of the embodiments of the present invention shown in Fig. 1, step a) is carried out. Step a) comprises dissolving lignin into an aqueous composition containing a compound selected from the class of phenols and alkali. The alkali is a hydroxide of an alkali metal. The temperature of the composition
15 is kept at 40 - 85 °C.

After the step of dissolving lignin, the composition formed in step a) is heated at a temperature, which is higher than the temperature of the composition in step a), with the proviso that the temperature
20 of the composition does not exceed 100 °C, while keeping the pH of the composition at a pH value of 6 - 14. During step b) the lignin is allowed to react with the compounds selected from the class of phenols and the alkali.

30 Step a) and step b) result in the lignin being suitable for further production steps and being suitable for being covalently bonded with e.g. formaldehyde.

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

The method illustrated in Fig. 2 begins with treating lignin in step a) and step b) in accordance with the method described above for Fig. 1.

After step b) the composition formed in step b) is mixed with a crosslinking agent in step c). The formed composition is heated at a temperature of 60 - 100 °C for allowing the reactant components, i.e. the lignin, the compound selected from the class of phenols and the crosslinking agent, in the composition to react with each other for forming a binder composition. The pH of the composition is kept, in step c), at a pH value of 6 - 14. The heating of the composition is continued until a desired, predetermined, viscosity value of the binder composition is reached.

As a result of step c) a binder composition having desired properties and especially having high ratio of bio-based components 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 - Activating lignin and the use of activated lignin for the production of a binder composition.

In this example lignin was firstly treated with phenol and sodium hydroxide. The following components and their amounts were used:

Material	MW	Concentration	Mass, g
NaOH	40	50 %	266
Water	18	0 %	500
Lignin	180	68.5 %	602
Phenol	94	90 %	458

Firstly lignin was mixed with water and then sodium hydroxide was added to the formed composition. The pH was increased to a value of about 13.8. To this

composition, the phenol was added. The temperature of the composition was kept at about 45 °C while dissolving lignin into the composition.

Then the temperature was increased to about
5 75 °C and the heating was continued for about 1 h. The pH of the composition was kept at a value of 10 - 11 during the heating of the composition.

Lignin treated as above described was thereafter used for producing a binder composition. 490 g
10 of formaldehyde was added to the above formed composition in a stepwise manner. The formed composition was heated at a temperature of about 83-85 °C and until the viscosity increased to 315 cP as measured at 25 °C using a rotary viscometer.

15 The formed binder composition was thereafter analyzed. The results of the analysis can be seen in Table 1.

Table 1. Analysis results

Specifications	
MR	1.9
Solids (2g, 2h at 120°C), %	42.3
Viscosity, cP	315
pH	12.1
Gel time 100 °C, min	48

20

In Table 1 MR stands for molecular ratio, i.e. the ratio of mol(formaldehyde)/mol(phenol + lignin).

The gel time was determined by adding 0.5 ml
25 of the sample in a test tube with a rod. The sample was not to be stirred. The test tube was heated to 100 °C in a glycerin or oil bath without mixing. The time until the sample became pasty (melted solid) was measured after which the measurement was completed.

30 From the results indicated in Table 1 it can be seen that the solids content is on a suitable level

for the binder composition to be used for wood panel products such as plywood or oriented strand boards.

EXAMPLE 2 - Activating lignin and the use of activated
5 lignin for the production of a binder composition.

In this example lignin was firstly treated with phenol and sodium hydroxide. The following components and their amounts were used:

10

Material	MW	Concentration	Mass, g
NaOH	40	50 %	66.0
Water	18	0 %	728.0
Lignin	180	70 %	879.0
Phenol	94	90 %	684.0

15

Firstly an aqueous composition was formed by mixing water, phenol and sodium hydroxide. To this aqueous composition, lignin was added. The temperature of the composition was kept at about 70 - 75 °C while dissolving lignin into the composition.

20

Then the temperature was increased to about 90 °C and the heating was continued for about 10 minutes. The pH of the composition was kept at a value of 8 - 8.3 during the heating of the composition.

25

Lignin treated as above described was thereafter used for producing a binder composition. 568 g of paraformaldehyde was added to the above formed composition in a stepwise manner. The formed composition was heated at a temperature of about 78 - 90 °C and until the viscosity was about 94 cP as measured at 25 °C using a rotary viscometer.

30

The formed binder composition was thereafter analyzed. The results of the analysis can be seen in Table 2.

Table 2. Analysis results

Specifications	
MR	1.9
Solids (2g, 2h at 120°C), %	52.8
Viscosity, cP	140
pH	7.6
Gel time 100 °C, min	85

In Table 2 MR stands for molecular ratio, i.e. the ratio of mol(paraformaldehyde)/mol(phenol + lignin).

5 The gel time was determined by adding 0.5 ml of the sample in a test tube with a rod. The sample was not to be stirred. The test tube was heated to 100 °C in a glycerin or oil bath without mixing. The time until the sample became pasty (melted solid) was measured after which the measurement was completed.

10 From the results indicated in Table 2 it can be seen that the solids content is on a suitable level for the binder composition to be used for laminates such as high-pressure laminates and for overlays. The water tolerance is better than for traditionally used phenol-formaldehyde binder compositions used for laminates such as high-pressure laminates and for shuttering films.

20 EXAMPLE 3 - Producing plywood

 The binder composition produced in Example 1 was used for the production of an adhesive composition. The binder composition was mixed with extenders, fillers, catalysts, additives, as examples of which e.g. starch, wood flour and hardener (e.g. tannin or carbonates) can be mentioned, thus forming the adhesive composition. Thereafter, wood veneers having the thickness of below 1.5 mm were glued together with the adhesive composition produced for producing a 9-plywood. After 8 min, cold-press panels were hot-

pressed at 130 °C. Results showed that the gluing effect was sufficiently good for gluing wood veneers.

EXAMPLE 4 - Producing shuttering films

5

The binder composition produced in Example 2 was used for the production of shuttering films. Kraft papers with a weight of 42 g/m² were impregnated with the binder composition and dried at 130 °C. The impregnated paper was pressed on one side of plywood at 145 °C for 6 min. Results showed that the shuttering film was performing sufficiently good; no blisters on the surface or delamination were observed.

15 EXAMPLE 5 - Activating lignin and the use of activated lignin for the production of a binder composition

In this example lignin was firstly treated with phenol and sodium hydroxide. The following components and their amounts were used:

Material	MW	Concentration	Mass, g
NaOH	40	50 %	66.0
Water	18	0 %	250.0
Lignin	180	70 %	879.0
Phenol	94	90 %	684.0

Firstly an aqueous composition was formed by mixing water, phenol and sodium hydroxide. To this aqueous composition, lignin was added. The temperature of the composition was kept at about 70 - 75 °C while dissolving lignin into the composition.

Then the temperature was increased to about 90 °C and the heating was continued for about 10 minutes. The pH of the composition was kept at a value of 8 - 8.3 during the heating of the composition.

Lignin treated as above described was thereafter used for producing a binder composition. 568 g of paraformaldehyde was added to the above formed composition in a stepwise manner. The formed composition was heated at a temperature of about 76 °C until the viscosity was about 930 cP as measured at 25 °C using a rotary viscometer.

The formed binder composition was thereafter analyzed. The results of the analysis can be seen in Table 3.

Table 3. Analysis results

Specifications	
MR	1.9
Solids (2g, 2h at 120°C), %	65.1
Viscosity, cP	780
pH	7.62
Gel time 130 °C, min	8

In Table 3 MR stands for molecular ratio, i.e. the ratio of mol(paraformaldehyde)/mol(phenol + lignin).

The gel time was determined by adding 0.5 ml of the sample in a test tube with a rod. The sample was not to be stirred. The test tube was heated to 130 °C in a glycerin or oil bath without mixing. The time until the sample became pasty (melted solid) was measured after which the measurement was completed.

The formed binder composition was used to produce a pre-preg by adding to the cooled binder composition firstly about 2 weight-%, based on the weight of the binder composition, of diethylene glycol (DEG) and then below 1 weight-%, based on the weight of the binder composition, of glycolic acid.

EXAMPLE 6 - Formation of rock wool

In this example rock wool was formed. Firstly lignin was treated with phenol and sodium hydroxide. The following components and their amounts were used:

Material	MW	Concentration	Mass, g
NaOH	40	50 %	111.7
Water	18	0 %	227.2
Lignin	180	70 %	171.9
Phenol	94	90 %	534.5

5

Firstly lignin was added to a composition of water, phenol and sodium hydroxide. To this composition 45.4 g of borax was added in a stepwise manner. Borax is a commonly used product in rock wool to enhance fire resistance and to act as an anti-moulding agent. Borax does not take part in the reactions where lignin is dissolved and allowed to react with phenol and NaOH. Borax can be added to the composition in the beginning of the process to ensure even distribution in the polymer matrix but it could also be added only after the binder composition formation.

10
15

The temperature of the composition was kept at about 42 °C and the pH of the composition was about 9 - 10 while allowing lignin to dissolve.

20

Thereafter the temperature of the composition was increased up to 50 °C and the lignin was allowed to react with phenol and sodium hydroxide in the composition for about an hour. The pH of the composition was about 9.3.

25

After the lignin had been activated by the above procedure, 1093.5 g of formaldehyde was added to the composition and the composition was heated at 65 °C for about 2.5 hours. The pH was kept at about 8.8. Then the composition was cooled to a temperature of 40 °C, about 1 weight-%, based on the weight of the composition, of boric acid was added followed by about 4

30

- 5 weight-%, based on the weight of the composition, of urea.

It is obvious to a person skilled in the art
5 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
limited to the examples described above; instead they
may vary within the scope of the claims.

What is claimed is:

1. A method for treating lignin, comprising:
 - a) dissolving lignin into an aqueous composition, which contains a compound selected from a class of phenols and alkali, while keeping a temperature of the composition between 40°C and 85°C and the pH of the composition at a pH value of between 4 and 10, wherein the alkali comprises a hydroxide of an alkali metal and wherein the compound selected from the class of phenols is selected from a group consisting of phenol, cresol, resorcinol and combinations thereof; and
 - b) heating the composition at a temperature, which is higher than the temperature of the composition in step a), with the proviso that the temperature of the composition does not exceed 100°C, while keeping the pH of the composition at a pH value of between 6 and 10, for allowing lignin to react with the compound selected from the class of phenols and the alkali.
2. The method of claim 1, wherein the temperature of the composition is kept, in step a), at a temperature between 65°C and 80°C.
3. The method of claim 1, wherein the temperature of the composition is kept, in step a), at a temperature between 70°C and 75°C.

4. The method of any one of claims 1 to 3, wherein step b) comprises heating the composition at the temperature, which is at least 5°C higher than the temperature of the composition in step a).

5. The method of any one of claims 1 to 3 wherein step b) comprises heating the composition at the temperature, which is at least 10°C higher than the temperature of the composition in step a).

6. The method of any one of claims 1 to 3 wherein step b) comprises heating the composition at the temperature, which is at least 20°C higher than the temperature of the composition in step a).

7. The method of any one of claims 1 to 6, wherein the pH of the composition is kept, in step a), at the pH value between 4.5 and 9.5.

8. The method of any one of claims 1 to 6, wherein the pH of the composition is kept, in step a), at the pH value between 5 and 9.

9. The method of any one of claims 1 to 6, wherein the pH of the composition is kept, in step a), at the pH value between 5.5 and 8.5.

10. The method of any one of claims 7 to 9, wherein the pH of the composition is kept, in step b), at the pH value between 6.5 and 9.5.

11. The method of any one of claims 7 to 9, wherein the pH of the composition is kept, in step b), at the pH value between 7 and 9.

12. The method of any one of claims 1 to 11, wherein the composition is heated, in step b), at the temperature between 60°C to 100°C.

13. The method of any one of claims 1 to 11, wherein the composition is heated, in step b), at the temperature between 70°C to 95°C.

14. The method of any one of claims 1 to 11, wherein the composition is heated, in step b), at the temperature between 75°C to 90°C.

15. A method for producing a binder composition, comprising:

a) dissolving lignin into an aqueous composition, which contains a compound selected from a class of phenols and alkali, while keeping a temperature of the composition at between 40°C and 85°C and a pH of the composition at a pH value of between 4 and 10, wherein the alkali comprises a hydroxide of an alkali metal and wherein the compound selected from the class of phenols is selected from a group consisting of phenol, cresol, resorcinol and combinations thereof;

b) heating the composition at a temperature, which is higher than the temperature of the composition in step a), with the proviso that the temperature of the composition does not

exceed 100°C, while keeping the pH of the composition at the pH value between 6 and 10, for allowing lignin to react with the compound selected from the class of phenols and the alkali; and

c) mixing the composition formed in step b) with a crosslinking agent and heating the composition at a temperature of between 60°C and 100°C while keeping the pH of the composition at the pH value between 6 and 10.

16. The method of claim 15, wherein step b) comprises heating the composition at a temperature of at least 5°C higher than the temperature of the composition in step a).

17. The method of claim 15, wherein step b) comprises heating the composition at a temperature of at least 10°C higher than the temperature of the composition in step a).

18. The method of claim 15, wherein step b) comprises heating the composition at a temperature of at least 20°C higher than the temperature of the composition in step a).

19. The method of any one of claims 15 to 18, wherein step c) comprises heating the composition at a temperature between 65°C and 95°C.

20. The method of any one of claims 15 to 18, wherein step c) comprises heating the composition at a temperature between 70°C and 90°C.

21. The method of any one of claims 15 to 18, wherein step c) comprises heating the composition at a temperature between 75°C and 85°C.

22. The method of any one of claims 15 to 21, wherein the pH of the composition is kept, in step a), at the pH value of between 4.5 to 9.5.

23. The method of any one of claims 15 to 21, wherein the pH of the composition is kept, in step a), at the pH value of between 5 to 9.

24. The method of any one of claims 15 to 21, wherein the pH of the composition is kept, in step a), at the pH value of between 5.8 to 8.5.

25. The method of any one of claims 22 to 24, wherein the pH of the composition is kept, in step b), at the pH value of between 6.5 and 9.5.

26. The method of any one of claims 22 to 24, wherein the pH of the composition is kept, in step b), at the pH value of between 7 and 9.

27. The method of any one of claims 15 to 26, wherein the pH of the composition is kept, in step c), at the pH value of between 6.5 and 9.5.

28. The method of any one of claims 15 to 26, wherein the pH of the composition is kept, in step c), at the pH value of between 7 and 9.

29. The method of any one of claims 15 to 28, wherein the crosslinking agent is an aldehyde.

30. The method of claim 29 wherein the crosslinking agent is at least one of formaldehyde and paraformaldehyde.

31. The use of lignin treated by the method of any one of claims 1 to 14 for the production of a binder composition.

32. A binder composition obtained by the method of any one of claims 15 to 30.

33. An adhesive composition comprising the binder composition of 32.

34. The use of a binder composition obtained by the method of any one of claims 15 and 25 to 28 for producing a laminate.

35. The use of claim 34 wherein the laminate is a high-pressure laminate.

36. The use of a binder composition obtained by the method of any one of claims 15 and 25 to 28 for producing one of a foundry material, a protective coating, a friction material, an abrasive material, glass wool, rock wool, a prepreg, a shuttering film, an overlay, a molding component and a fiber reinforced composite.

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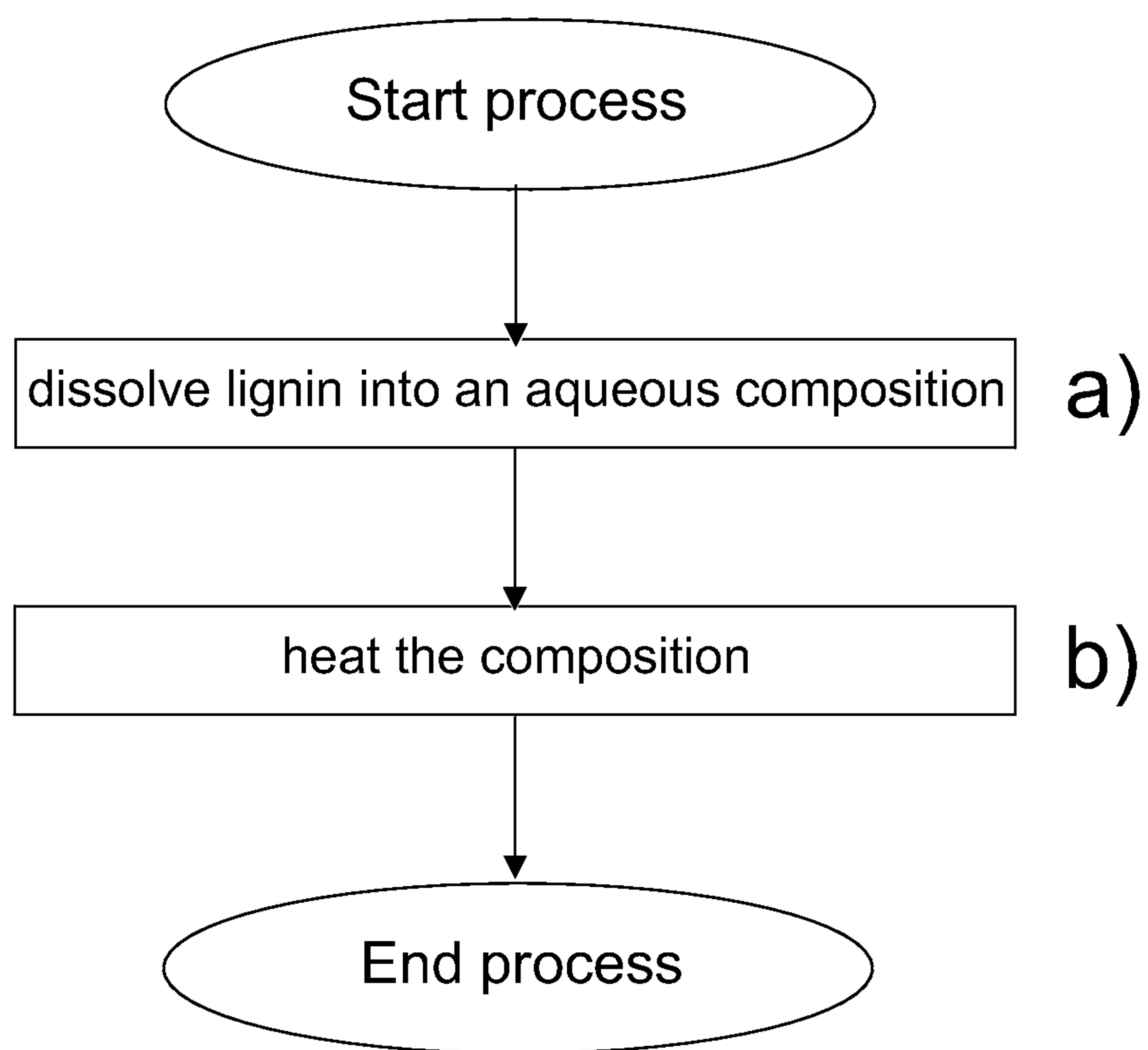


Fig. 1

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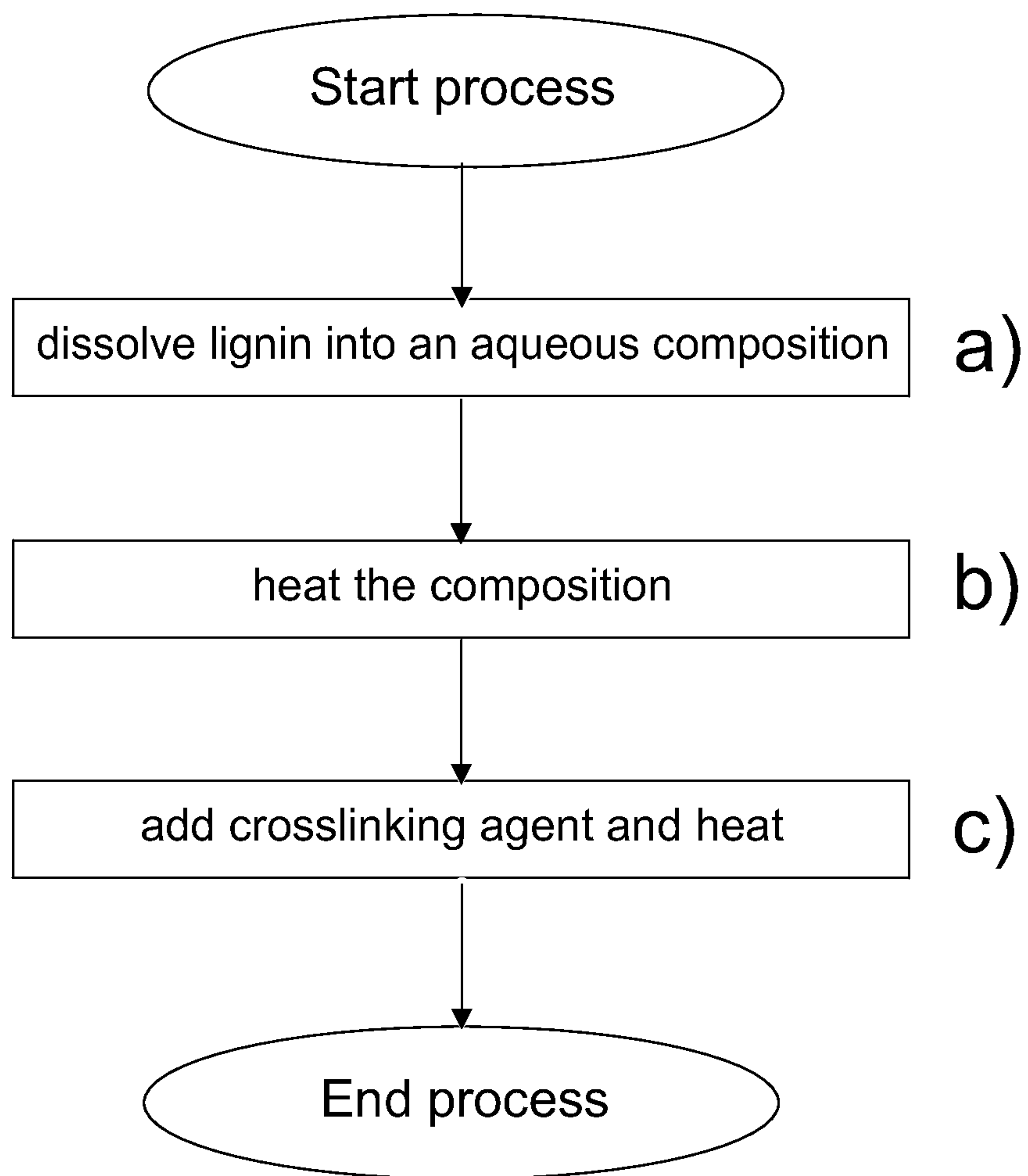


Fig. 2