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(54) Title: MODIFIED LIGNINS

(57) **Abrégé/Abstract:**

Ligning and polyether glycol produces a product which has a distinct melt point and melt viscosity. This has a plasticizing effect wherein the polyether glycol can be thought of as the hard resin. When 20 parts of polyether glycol are processed with 80 parts of lignin a new material which has a melt point and a viscosity is created whereas lignin itself has no melt and flow characteristics. The new modified lignin can be cured with hexamine like a phenolic resin.



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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US97/15039 <b>(22) International Filing Date:</b> 26 August 1997 (26.08.97)  <b>(30) Priority Data:</b> 60/024,558                      26 August 1996 (26.08.96)                      US  <b>(71)(72) Applicant and Inventor:</b> KURPLE, Kenneth, R. [US/US]; 9533 Springborn Road, Anchorville, MI 48004 (US).  <b>(74) Agents:</b> MITCHELL, Kenneth, L. et al.; Woodling, Krost & Rust, Suite 1520, 1801 East Ninth Street, Cleveland, OH 44114 (US).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, HU, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> MODIFIED LIGNINS  <b>(57) Abstract</b> <p>Ligning and polyether glycol produces a product which has a distinct melt point and melt viscosity. This has a plasticizing effect wherein the polyether glycol can be thought of as the hard resin. When 20 parts of polyether glycol are processed with 80 parts of lignin a new material which has a melt point and a viscosity is created whereas lignin itself has no melt and flow characteristics. The new modified lignin can be cured with hexamine like a phenolic resin.</p>		

### MODIFIED LIGNINS

There are substantial markets available to lignins if they can achieve the same melt and flow characteristics as a phenolic resin. There are at least two major factors of this present invention which allow these new modified lignins to be able to totally replace phenolic resins. The first factor that has been that a lignin which does not have any distinct melt point or flow characteristics can be modified with a certain amount of polyether glycol to achieve a certain desired melt point. So, what occurs is a certain ratio of lignin to a polyether compound (such as polyether glycol) produces a product which has a distinct melt point and melt viscosity. This effect can be thought of as a type of plasticizing effect wherein the lignin can be thought of as the hard resin. For example, when 20- parts of a polyether glycol is properly processed with 80 parts of lignin what results is a new material which now has melt and flow characteristics, whereas lignin by itself has no melt and flow characteristics. Because these melt and flow characteristics are similar to a phenolic resin these new modified lignins can be cured with hexamine just like a phenolic resin. This is because the lignin molecule contains phenolic groups which are reactive to hexamine just as in a phenolic resin. This is significant because although lignin itself does contain phenolic groups that are reactive to hexamine the fact that the lignin molecule does not melt and flow means that it would be almost impossible to obtain a commercially moldable part from a press with heat and pressure.

Another major advantage of this present invention is that lignin is derived from wood, which is a natural product. As in all natural products there will exist a certain amount of variation depending on the exact type of wood, age and other conditions. These variations are another major reason why lignins have not been extensively utilized as replacements for phenolic resins, whereas a synthetic phenolic resin can be made to exact and



reproducible specifications, and the lignin molecule may have wide variations. Therefore, if there is a variation in the lignin this can be adjusted by changing the ratio of lignin to a polyether compound (such as polyether glycol) in order to achieve the desired melt and flow characteristics.

Another aspect of this invention that has a synergistic effect on the melt and flow characteristics in the sodium level of the lignin. It was discovered that if the sodium level is too high the modified lignin will not have melt and flow characteristics which can be utilized as a commercial product. It was found that the same lignin, but with different sodium levels, will produce modified lignins with wide variations in melt and flow characteristics. Therefore, it was discovered that there is a certain critical level for the presence of sodium and other ionic materials above which no useful products could be obtained. This critical level may vary depending on the specific nature or chemical composition of the lignins. In general, experimentation has determined that the sodium level of lignin should preferably be less than 22 percent by weight of sodium.

Another major invention is the fact that lignins are a natural product and as such there may be natural chemical variations in the lignin molecule. These variations may include different functional groups or different concentrations of these functional groups. These variations become rather significant when you are trying to formulate a product that must have, in addition to certain physical properties such as melt and flow, certain curing characteristics in order to produce a consistent product day in and day out.

If there is a variation in the lignin, then this variation can be accommodated by changing the ratio of lignin to a polyether compound (such as polyether glycol) in order to achieve the desired melt and flow characteristics. Preferably, the ratio of the polyether compound to lignin is from 1 percent to 70 percent. The polyether compound can be made from the reaction of propylene oxide or ethylene oxide with alcohols, amines, nonylphenol, phenol or any phenolic compound.

These curing characteristics are very important because in addition to the physical properties of the product they also determine the productivity, which directly affects how profitable the product will be. One major market for lignins is to either partially or totally replace phenolic resins in many products that currently use all phenolic resins. There are at least two major advantages why lignins can be used as either partial or total replacement of phenolic resins in phenolic production. One major advantage is that lignins can offer significant economic advantages. Since most phenolic resins are made from phenol and phenol is a petrochemical product, cost of phenol is very dependent on the cost of oil. So, as the cost of oil increases, the cost of phenol and phenolic resins will also increase. These price increases could make phenolic resins less competitive and thereby make it possible for these phenolic products to be replaced by other products. Therefore, when more lignin can be incorporated into a phenolic product, this could offset price increases and make these lignin modified phenolic products more profitable and more competitive, thus increasing their market share.

#### EXAMPLE I

In a suitable vessel 20 parts of Pluracol P-410™ is heated to 200°F and 80 parts of lignin is slowly added while adequate agitation is maintained and the heat slowly increased to 200°F. The viscosity of this mixture will increase significantly but it still is able to be mixed. Then once all of the lignin is added, the mixture is continued to be mixed until everything is homogeneous. Then this mixture is poured out on to a surface which allows this mixture to cool and form a hard brittle material. This lignin is now in a form which is very similar to phenolic resins. This modification of the lignin now makes it possible for it to have melt and flow characteristics.



What is claimed is:

1. A composition comprising lignin blended with a polyether compound to improve the melt and flow characteristics of the lignin, said lignin having a sodium level less than 22% by weight of sodium.
2. A composition as claimed in claim 1 wherein the percentage of said polyether compound is from 1% to 70% of the composition.
3. A composition comprising lignin blended with a polyester compound to improve the melt and flow characteristics of the lignin, said lignin having a sodium level less than 22% by weight of sodium.
4. A composition as claimed in claim 3 wherein the percentage of said polyester compound is from 1% to 70% of the composition.
5. A composition as claimed in claim 1 wherein said polyether compound is produced by a reaction involving at least one of ethylene oxide and propylene oxide.
6. A composition comprising lignin and blended with a combination of polyester resins and polyether glycols to improve the melt and flow characteristics of the lignin, said lignin having a sodium level less than 22% by weight of sodium.
7. A composition comprising a polyether derivative (derived from at least one of ethylene oxide and propylene oxide) which is initiated by a chemical selected from the group consisting of nonlyphenol, phenol and a substituted phenol blended with lignin to improve the melt and flow characteristics of lignin, said lignin having a sodium level

less than 22% by weight of sodium.

8. A composition as claimed in claim 1 wherein the modified lignin is reacted with hexamine to produce a cured product.

9. A composition as claimed in claim 3 wherein the modified lignin is reacted with hexamine to produce a cured product.

10. A composition as claimed in claim 6 wherein the modified lignin is reacted with hexamine to produce a cured product.

11. A composition as claimed in claim 7 wherein the modified lignin is reacted with hexamine to produce a cured product.