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METHOD OF IMPROVING THE PROCESSING OF REFINED CHEMICAL PULP INTO VISCOSE

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14 Claims. (Cl. 260—217)

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This invention relates to the production of cellulosic products, and has for its general object the provision of certain improvements in carrying out one or more of the processing steps in the production of such products. The invention is of special advantage in the preparation and processing of viscose derived from chemically prepared wood pulp, and in this connection aims to improve the steps of shredding and filtering. The invention provides a method of processing chemical wood pulp comprising the incorporation of an aliphatic alcohol with from 7 to 18 carbon atoms in the pulp prior to the completion of shredding. The invention also provides, as a new article of manufacture, a chemically prepared wood pulp product having incorporated therein an aliphatic alcohol with from 7 to 18 carbon atoms.

Chemically prepared wood pulps are extensively used in the industrial arts for the production of such cellulosic products as rayon and other synthetic fibers, nitrocellulose, cellulose acetate, cellulose ethers, Cellophane and similar cellulosic films or sheets, etc. Wood pulp is commonly prepared and marketed in sheet form, and comminution or shredding is customarily one of the initial steps in its subsequent processing. When wood pulp is used in bulk form, either wet or dry, shredding or comminution is frequently a step involved in the subsequent processing. Since such subsequent processing usually involves the conversion of the cellulose of the wood pulp to some cellulose derivative and the solution of the derivative in a suitable solvent, the purpose of shredding is to break down the sheet into a fluffy mass or crumb in which the individual fibers will be suitably exposed to the action of the derivative-forming reagent or reagents. Thus, for example, in the production of viscose, the wood pulp sheet is customarily steeped in a caustic soda solution to form alkali cellulose, and the alkali cellulose, after pressing and while still in sheet form, is shredded or comminuted to properly prepare the cellulose fibers for the xanthating reaction with carbon bisulfide. The resulting sodium cellulose xanthate is dissolved in dilute caustic soda to form the solution commonly known as viscose, which is opacified if desired, filtered, and spun into filaments.

Normal dissolving wood pulps in present use consist mainly of cellulose, but contain appreciable amounts of noncellulosic impurities, such as hemicelluloses, fats, resins, waxes, etc. One of the main objects in the manufacture of a highly refined dissolving pulp is to remove as much as

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possible of the noncellulosic impurities, so that a whiter, purer pulp results, which is capable, in the manufacture of rayon, of producing a higher grade yarn.

We find, however, that not all of the noncellulosic impurities which can be removed are undesirable, and in fact, certain of such impurities, normally present in small amounts, are highly beneficial in aiding the processing of the pulp into viscose, especially as regards the step of shredding the alkali cellulose. The beneficial impurities which aid the shredding operation are probably of the nature of fiber lubricants which permit the steeped and pressed pulp to be thoroughly shredded into a fluffy condition more readily and without mechanical damage to the alkali cellulose fibers, which would cause them to react incompletely with carbon bisulfide. In a pulp which has not been highly refined, most of these beneficial impurities constitute a portion of the materials removable by organic solvents, as for example, ether, benzene, alcohol, etc. These beneficial impurities, often loosely termed "resins," are mainly of the nature of waxes, fats and resins, the latter often being present in a relatively small amount.

In theory, the problem of making a good pulp could be solved by removing all the undesirable impurities while retaining those impurities which facilitate the shredding operation by lubricating the fibers or otherwise. In practice, such a clean-cut separation is difficult to accomplish directly. We have discovered that better results are obtainable by removing most or all of the impurities, including those which serve beneficially as lubricants to the alkali cellulose fibers during shredding, and then adding to the pulp or to the alkali cellulose prior to the completion of shredding a sufficient amount of a material of a class entirely different from the natural impurities originally present, and which greatly aids the steps of shredding and filtering.

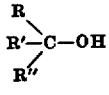
White, highly purified or refined wood pulps are very advantageous for the production of high grade rayon yarns of superior strength and color, and for this reason, are highly esteemed by the trade. Such highly refined pulps are in general characterized by having an ether extract of about 0.15% or less where such values refer to the amount of natural ether-extractable material left in the pulp after the purification processes. When the pulp is to be processed into viscose by normal rayon processing conditions, our invention is particularly applicable to such highly refined pulps containing not more than about 0.15%

of ether-extractable material, and increasingly so as the ether extract approaches zero. It may be applied with marked advantages to the processing of dissolving pulps containing up to 0.30% of ether-extractable material, when processing conditions are used involving the pressing of the steeped alkali cellulose to low press ratios and xanthating with minimum or near minimum amounts of carbon bisulphide. This is of especial importance in the manufacture of such products as transparent film and staple fiber by the viscose process.

The surface-active portion of the beneficial impurities, naturally present as such in the refined pulps or formed during the processing, are predominately of the general class known as anion-active materials.

We have discovered that a class of nonionic compounds, namely aliphatic alcohols with 7 to 18 carbon atoms, give important results in the processing of chemical pulp. In general, the compounds which we have discovered for use in our invention are more effective and more stable in regard to deterioration by ageing than the beneficial portion of the naturally occurring impurities. This property becomes important in those cases where wood pulp subject to such deterioration must be stored over a considerable period of time. Based on these discoveries, our invention involves improving the inherent properties of the wood pulp as well as one or more of the processing steps in the production of cellulosic products from chemically prepared wood pulp by carrying out one or more of the processing steps in the presence of an aliphatic alcohol with from 7 to 18 carbon atoms. When applied to the preparation and processing of viscose derived from chemically prepared wood pulp, the invention particularly involves carrying out the shredding of the alkali cellulose, or at least the final stage of shredding, and the filtration of the viscose in the presence of the aliphatic alcohol.

Aliphatic alcohols are considered as that class of organic compounds comprising the mono hydroxyl derivatives of the paraffin hydrocarbons. A general structural formula covering all aliphatic alcohols is



where the substituents as represented by R, R', and R'' may be any aliphatic radicals or hydrogen. If two of the substituents are hydrogen, then the alcohol is considered a primary alcohol. One hydrogen on the carbon atom gives a secondary alcohol while the presence of no hydrogens on the carbon atom results in a tertiary alcohol. Certain members of these three types of alcohols are effective in the practice of the invention.

The compounds which we have found to be effective and suitable in the practice of our invention are the aliphatic alcohols, either primary, secondary or tertiary saturated or unsaturated and having from 7–18 carbon atoms. The most preferred class, with regard to both effectiveness and stability in ageing of the pulp, are the saturated aliphatic alcohols within the range of 7–18 carbon atoms and melting below 25° C.

All of the alcohols of the invention are sufficiently non-volatile to remain in the pulp so treated over long periods of time. The lower numbers of the series do, however, possess sufficient vapor pressure so that they impart to pulp treated with them a pronounced odor. Thus,

where lack of odor is a consideration, it will usually in practice be preferred to use those members within the series having at least 10 carbon atoms.

Among the alcohols which we have used successively in practicing our invention are the following: heptadecanol (3,9-diethyltridecanol-6), tetradecanol (7-ethyl-2-methylundecanol-4), undecanol (5-ethylnonanol-2), n-lauryl alcohol, decanol, and octyl alcohol (2-ethyl hexanol). Three of the compounds which we have found to be most effective are 7-ethyl-2-methylundecanol-4, n-lauryl alcohol and octyl alcohol.

All of the compounds listed above are commercially available and technical grades function in essentially the same manner as the pure compounds.

The application of the compounds to the pulp may be accomplished by use of a solution of the compound in a satisfactory organic solvent but more practically by use of an aqueous emulsion. The preparation of such an aqueous emulsion may be effected by any appropriate procedure common to the art, but our preferred method is to dissolve the alcohol in ordinary commercial grade ethyl alcohol (forming a 5 to 15% solution) and then to pour this solution into a large volume of water containing about 5% (based on the weight of the added substantially non-volatile alcohol) of an alkyl aryl sulphonate such as "Santomerse No. 3" or other suitable emulsifying agent. A suspension or emulsion so prepared is sufficiently stable for application to a pulp sheet and may be applied by spraying or by means of a rotating roll partly immersed in the emulsion, as described elsewhere in more detail.

For application during the shredding operation, the compounds may be sprayed directly onto the alkali cellulose as a concentrated solution in a suitable organic solvent—preferably a water miscible, volatile solvent such as ethyl alcohol or isopropanol. The compounds may also be sprayed or otherwise distributed on the alkali cellulose without the use of solvents in their normal liquid or melted states. We have discovered that esters of the alcohols of the invention which decompose in the presence of caustic soda may be likewise used since they liberate the compounds of the invention during the steeping and ageing periods and thus in effect add them to the alkali cellulose prior to the completion of shredding.

Octyl alcohol (2-ethyl hexanol) is one of the most effective agents of the invention. When applied to the pulp as such, however, it has the disadvantage that the pulp treated with it has a noticeable odor during storage. Thus, as regards the application of octyl alcohol, we prefer to add it in the form of the dioctyl ester of sodium sulphosuccinate ("Aerosol OT") in which form it has the advantage of being water soluble and non-odoriferous. This compound then decomposes either largely or wholly during the steeping and ageing periods and thus in effect adds octyl alcohol (2-ethyl hexanol) to the alkali cellulose prior to the completion of shredding. The other alcohols of the invention may be also similarly applied using water soluble esters of sodium sulphosuccinate or other innocuous compounds.

While highly refined wood pulps are advantageous for the production of high grade rayon yarns of superior strength and color, the reaction of the shredded alkali cellulose from such pulps with carbon bisulfide is frequently incomplete. This impairment in the xanthating activity of

the shredded alkali cellulose is due to some damage to the fibers during the shredding operation or to incomplete comminution or to formation of compressed fiber bundles. In the case of the conventional shredder having revolving blades coacting with a stationary saddle bar, the tendency for such damage is especially great if the clearance between the revolving blades and the saddle bar is a little less than the correct value. In an extreme case, with a very highly refined pulp, shredding, even under optimum conditions, may produce a shredded pulp which xanthates less completely than if the alkali cellulose were not shredded at all. In other cases, it is possible to obtain reasonably satisfactory shredding of the alkali cellulose from highly refined pulps by adjusting the shredder clearance and by experimenting to find the optimum shredding time for the particular pulp and particular shredder. In this manner it is possible in some cases to obtain almost as complete a degree of xanthation of the shredded alkali cellulose from a highly refined pulp as would be obtained with the shredded alkali cellulose from a less pure pulp. But the necessity of constant supervision and adjustment to assure optimum shredding conditions makes the shredding of highly refined pulps too sensitive for satisfactory commercial practice. Furthermore, even with the most careful supervision, such very pure pulps would not give satisfactory filtering viscose solutions in the event of using low press ratios and near minimum amounts of carbon bisulphide. The shredding of the alkali cellulose from highly refined pulps, or even from less pure pulp in the event of using low press ratios and near minimum amounts of CS_2, is improved, in our invention, by carrying out the shredding operation in the presence of an aliphatic alcohol of 7–18 carbon atoms which may be added to the pulp prior to use in the viscose process or to the alkali cellulose prior to the completion of shredding. The improvement in shredding produces a shredded alkali cellulose which reacts substantially completely with carbon bisulfide, and the resulting viscose is comparatively free of unreacted fibers and filtration proceeds more rapidly and economically.

In the usual viscose process the sheets are first subjected to a steeping step to convert the cellulose to alkali cellulose, and the pressed sheets of alkali cellulose are then shredded to form a fluffy mass of fibers. (By "normal" press ratios in this application are meant ratios of pressed alkali cellulose to air dry pulp of about 2.8–3.0 or higher. By "low" ratios are meant values of about 2.8–2.5 or lower, and especially those not exceeding 2.7.) The fluffy mass is xanthated, dissolved in dilute causic soda and the solution commonly known as viscose filtered to remove undissolved fibers and gel-like materials, and ripened to impart the desired properties for satisfactory spinning.

The most practical and economical manner of securing the desired presence of the aliphatic alcohols of our invention during the shredding of the alkali cellulose and during the filtration of the viscose is to incorporate the aliphatic alcohol in the wood pulp. This may be advantageously effected by adding the aliphatic alcohol to the pulp on the sheet-forming machine subsequent to sheet formation but prior to complete drying, by spraying the pulp sheet with an aqueous emulsion of the compound or by means of a rotating roll partly immersed in such an emulsion. If desired, the aliphatic alcohols of the

invention may be incorporated in the pulp prior to sheet formation, as for example, by adding the agent to a suspension of pulp in water in a stock chest. In the latter case, the white water may in some instances be advantageously recirculated in order to prevent loss of the agent when eliminating water in sheet formation. This, however, will not always be necessary. In any case, there is produced a chemically prepared wood pulp product having an aliphatic alcohol with from 7–18 carbon atoms incorporated therein. When the aliphatic alcohol is so incorporated in the wood pulp, by the manufacturer thereof, the pulp comes to the rayon manufacturer in a form calculated to secure the full advantages of the invention in the preparation and processing of the viscose into rayon yarns, transparent films, staple fiber or the like.

The amount of the aliphatic alcohols used in the practice of the invention is relatively small, ranging from 0.01 to 0.25%, and preferably from 0.02 to 0.10%, by weight on the bone dry weight of the wood pulp used, when the agent is incorporated in or added to the pulp or to the alkali cellulose. So far as the objectives of the invention are concerned, there is little if any improvement by increasing the amount of aliphatic alcohol above 0.25% and such higher amounts frequently give rise to certain disadvantages. These disadvantages include the causing of excessive softness in the sheet, resulting in mechanical difficulties in steeping, excessive ball formation in xanthation, difficulties in the dissolving operation due both to the excessive ball formation in the xanthating step and due to excessive foaming in the viscose solution. Also there may be considerable difficulty in obtaining a completely deaerated viscose which is absolutely necessary for satisfactory spinning. Higher concentrations of the aliphatic alcohols may also cause excessive turbidity of the viscose and may even cause poor filtration by separation of the aliphatic alcohol from the viscose. Also in the event of using unduly high proportions of the aliphatic alcohols the final regenerated cellulose product will tend to be "delustered" and of lower strength.

While it is our preferred practice to incorporate the aliphatic alcohol in the wood pulp, more particularly refined pulp containing not more than about 0.30% of ether-extractable material, and for rayon purposes of rayon yarn preferably not more than 0.15% of ether-extractable matter, the presence of the compound during the processing steps of shredding may be secured in any other appropriate manner. Moreover, when the pulp is in sheet form, the compound need not be incorporated in every sheet, but may be incorporated in only alternate sheets, or otherwise distributed throughout the pulp. It may be added to only a portion of the pulp in whatever form it is marketed. Alternatively, the compound may be sprayed upon or otherwise suitably added to all or a portion of the alkali cellulose prior to shredding or prior to the completion of shredding. However, we believe it will generally be found more advantageous to incorporate the aliphatic alcohol in the initial wood pulp product, both as a matter of convenience and economy in preparing and processing the viscose, and because a very uniform distribution of the compound during shredding and in the final viscose is easily attained.

When the invention is practiced for effecting the hereinbefore-mentioned improvements in

shredding, certain further economies are effected in the subsequent steps of xanthating, dissolving, and filtering. In viscose solutions there is usually a certain amount of undissolved fibers and gel-like material due to the incomplete reaction of the cellulose with the carbon bisulfide during xanthation. Prior to spinning, the viscose solutions are filtered several times to remove these gels and undissolved fibers. In the event that the viscose solutions contain excessive amounts of undissolved and partially dissolved fibers, filtration is an expensive operation. In such cases the filters become rapidly clogged and the filter media must be changed frequently in order that the viscose will pass through in a reasonable time. Frequent changing of the filter media is expensive, not only as regards consumption of filter cloth but also in view of the very considerable amount of labor involved and also since a certain amount of viscose is lost every time the filter is opened. Furthermore, when the viscose solutions contain very large proportions of gel-like material, filtration is usually not altogether satisfactory in that some of the smaller gel-like particles tend to pass through the pores of the cloth with adverse effect upon the spinning operation. It has heretofore been the practice in the viscose industry, when processing pulps which tend to yield viscose solutions high in undissolved material and having poor filtration properties, to minimize such difficulties by carrying out the xanthation with amounts of carbon bisulfide considerably in excess of that normally required. Use of excess carbon bisulfide is expensive and in addition is technically undesirable in that it may adversely affect certain properties such as the ripening of the viscose and yarn characteristics. We have found that when processing purified pulps which would normally tend to give poorly filtering viscose solutions, the addition of minute amounts of the aliphatic alcohols of the invention so improves the shredding operation that the alkali cellulose subsequently reacts much more completely with carbon bisulfide and yields a viscose solution very free from undissolved and partially dissolved cellulose particles and having good filtration properties. This result can be accomplished not only without the use of excess carbon bisulfide, but in many cases satisfactory viscose solutions can be obtained using amounts of carbon bisulfide very substantially less than the amounts normally required.

We claim:

1. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to one of the viscose-forming materials at a stage prior to completion of shredding an aliphatic alcohol having from 7–18 carbon atoms, said alcohol being added in a range from 0.01% to 0.25% by weight, such percentages being based on the weight of the bone dry pulp.

2. The method of improving the manufacture of shredded alkali cellulose from refined chemical pulp containing not more than 0.30% ether-extractable matter comprising adding to one of the alkali cellulose-forming materials at a stage prior to the completion of shredding an aliphatic alcohol having from 7–18 carbon atoms, said alcohol being added in a range from 0.01% to 0.25% by weight, such percentages being based on the weight of the bone dry pulp.

3. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to such refined chemical pulp prior to use in the viscose process an aliphatic alcohol having from 7–18 carbon atoms, said alcohol being added in a range from 0.01% to 0.25% by weight, such percentages being based on the bone dry weight of the pulp.

4. As a new article of manufacture, a refined chemical pulp product containing not more than 0.30% of natural ether-extractable matter and having incorporated therein an aliphatic alcohol having from 7–18 carbon atoms, the amount of the incorporated alcohol being from 0.01% to 0.25% by weight, such percentages being based on the bone dry weight of the pulp.

5. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to one of the viscose-forming materials at a stage prior to completion of shredding a saturated aliphatic alcohol within the range of 7–18 carbon atoms and melting below 25° C., said aliphatic alcohol being added in a range from 0.01% to 0.25% by weight, such percentages being based on the weight of the bone dry pulp.

6. The method of improving the manufacture of shredded alkali cellulose from refined chemical pulp containing not more than 0.30% ether-extractable matter comprising adding to one of the alkali cellulose forming materials at a stage prior to the completion of shredding a saturated aliphatic alcohol within the range of 7–18 carbon atoms and melting below 25° C., said alcohol being added in a range from 0.01% to 0.25%, such percentages being based on the weight of the bone dry pulp.

7. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to such refined chemical pulp prior to use in the viscose process a saturated aliphatic alcohol within the range of 7–18 carbon atoms and melting below 25° C., said alcohol being added in a range from 0.01% to 0.25% by weight, such percentages being based on the bone dry weight of the pulp.

8. As a new article of manufacture, a refined chemical pulp product containing not more than 0.30% of natural ether-extractable matter and having incorporated therein a saturated aliphatic alcohol within the range of 7–18 carbon atoms and melting below 25° C., the amount of the incorporated alcohol being from 0.01% to 0.25% by weight on the bone dry pulp.

9. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to one of the viscose-forming materials at a stage prior to completion of shredding n-lauryl alcohol in a range from 0.01% to 0.25% by weight, such percentages being based on the weight of the bone dry pulp.

10. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to such refined chemical pulp prior to use in the viscose process n-lauryl alcohol in a range from 0.01% to 0.25% by weight, such percentages being based on the bone dry weight of the pulp.

11. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to one of the viscose-forming materials at a stage prior to completion of shred-

ding 7-ethyl-2-methylundecanol-4 in a range from 0.01% to 0.25% by weight, such percentages being based on the weight of the bone dry pulp.

12. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to such refined chemical pulp prior to use in the viscose process 7-ethyl-2-methylundecanol-4 in a range from 0.01% to 0.25% by weight, such percentages being based on the bone dry weight of the pulp.

13. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to one of the viscose-forming materials at a stage prior to completion of shredding 2-ethyl hexanol in a range from 0.01% to 0.25% by weight, such percentages being based on the weight of the bone dry pulp.

14. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to such refined chemical pulp prior to use in the viscose process 2-ethyl hexanol in a range from 0.01% to 0.25% by weight, such percentages being based on the bone dry weight of the pulp.

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REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number	Name	Date
2,331,936	Schlosser et al.	Oct. 19, 1943
1,842,689	Richter	Jan. 26, 1932
2,166,314	Martin	July 18, 1939
2,286,962	Hegan et al.	June 12, 1942
2,083,744	Richter	June 15, 1937
1,986,291	Schur	Jan. 1, 1935
2,085,709	Steibelt	June 29, 1937

Certificate of Correction

Patent No. 2,432,129. December 9, 1947.

PAUL HENRY SCHLOSSER ET AL.

It is hereby certified that error appears in the printed specification of the above numbered patent requiring correction as follows: Column 4, lines 5 and 6, for the word "successively" read *successfully*; and that the said Letters Patent should be read with this correction therein that the same many conform to the record of the case in the Patent Office.

Signed and sealed this 16th day of March, A. D. 1948.

[SEAL]

THOMAS F. MURPHY,
Assistant Commissioner of Patents.

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13. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to one of the viscose-forming materials at a stage prior to completion of shredding 2-ethyl hexanol in a range from 0.01% to 0.25% by weight, such percentages being based on the weight of the bone dry pulp.

14. The method of improving the processing of refined chemical pulp containing not more than 0.30% ether-extractable matter into viscose comprising adding to such refined chemical pulp prior to use in the viscose process 2-ethyl hexanol in a range from 0.01% to 0.25% by weight, such percentages being based on the bone dry weight of the pulp.

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