Synthesis of Pulping Processes With Fiber Loading Methods for Lightweight Papers

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ABSTRACT

Pulping technologies can be synthesized with fiber loading with simultaneous alkaline peroxide bleaching to produce lightweight high-opacity printing papers. We compared the results of recent experiments on combining oxalic acid pretreated wood chips used for thermomechanical pulp (TMP) with fiber loading and previous experiments on combining similar pulps treated with fungal and microwave pretreatments with fiber loading. For TMP at 100 mL CSF, various species and pretreatments resulted in differences in (a) energy requirements for pulp production, (b) properties of handsheets prepared from pulps, and (c) response of pulps to precipitated calcium carbonate added conventionally and by fiber loading. Of the three treatment methods (oxalic acid, fungal (bio-treatment), and microwave), refining energy reduction was greatest for bio-treated TMP (31%). For microwave and oxalic acid treated pulps, energy was reduced 10% and 20%, respectively. All three pretreatments improved handsheet strength properties to about the same extent. Brightness and scattering coefficient were reduced the least by oxalic acid treatment, followed by microwave and bio-pretreatments. Fiber loading of bio-pretreated TMP improved handsheet strength compared with that of control TMP. Fiber loading and direct loading of oxalic acid pretreated pulps resulted in similar handsheet strengths.

INTRODUCTION

The forest products industry is among the nine most energy-intensive U.S. industries identified by the Office of Industrial Technologies of the Department of Energy (DOE). The forest products industry employs more than 1.3 million people and ranks among the top industries in 46 States. Agenda 2020, the forest and paper industry's vision for the future, in partnership with the DOE, proposes the development of new methods or technologies to reduce energy consumption, enhance economic competitiveness, and reduce environmental impact.

Mechanical pulping accounts for about 25% of wood pulp production worldwide. This volume is expected to increase in the future, because mechanical pulping has a high yield and is used to extend forest resources. Current efforts in mechanical pulping are directed towards producing lightweight papers. When grammage of printing paper is decreased, mechanical pulps are typically used to maintain bulk and scattering coefficient. However, the strength of the paper is usually decreased. To reduce grammage and maintain acceptable paper performance, paper optical properties and paper strength must be increased.

The Forest Products Laboratory (FPL) has developed a number of technologies that can be used in combination or synthesized to produce lightweight high-opacity printing papers. These technologies include biopulping (1–8), microwave pretreatment prior to TMP pulping (9), oxalic acid pretreatment pulping, and fiber loading (10–17) with simultaneous alkaline peroxide bleaching (18).

Biopulping, defined as the fungal treatment of wood chips prior to mechanical pulping, enhances the strength properties of the paper and reduces the consumption of mechanical pulping energy. Mechanical pulping is electrical-energy-intensive and yields paper with less strength. A mill that processes 300 tons of pulp per day spends about $10 million annually in electricity costs. Biopulping overcomes both energy use and pulp strength problems while reducing the toxicity of the waste stream from mechanical pulping.

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During biopulping, chips are inoculated with white-rot fungi, which colonize the surface and interior of the chips and secrete lignin-degrading enzymes that modify and soften the wood. The resultant fibers are longer, more highly fibrillated, and more porous than fibers from untreated chips. Increased fiber length and fibrillation improves interfiber bonding and corresponding paper strength.

Microwave processing of logs can reduce the total energy required for producing mechanical pulp while increasing the fiber bonding strength of the handsheets (9). Preliminary experiments have shown that the total electrical energy required for mechanical pulping can be reduced by 15% if the logs are first microwaved. Additionally, the strength properties of the resulting handsheets can be increased by as much as 15% to 30%. Microwave equipment is cost effective; with the resulting benefits from lower energy use and stronger paper, microwave pretreatment may lead to wider use of mechanical pulping. Oxalic acid pretreatment of wood chips prior to mechanical pulping has been shown to reduce pulping energy at least 25% while producing a stronger paper.

Unfortunately, all of these processes (biopulping, microwaving, and oxalic acid pretreatment) reduce the brightness and opacity of the resulting paper. High brightness and opacity are absolutely critical for the production of high quality papers, particularly when the trend in the pulp and paper industry is towards producing lightweight paper. Fiber loading is a technology in which calcium carbonate filler is deposited onto the surface and inside the fibers, which increases the brightness and opacity of papers significantly. Fiber loading therefore has the potential to solve both the brightness and opacity problems associated with biopulping, microwaving, and oxalic acid pretreatment.

Simultaneous alkaline peroxide bleaching and fiber loading eliminates the use of sodium hydroxide (17) in TMP bleaching. By eliminating sodium hydroxide and replacing it with calcium hydroxide (needed for fiber loading), manufacturing costs are reduced, as well as the COD content of process water. Moreover, this combined process increases the strength of filled papers made through the biopulping process, reducing the need for chemical reinforcing pulps in printing and writing papers.

In this report, we compare and contrast the results of recent experiments using TMP from oxalic acid pretreated wood chips with past results from experiments with similar pulps prepared using biopulping and microwave pretreatments along with fiber loading of bio pre-treated and oxalic acid pre-treated TMPs.

EXPERIMENTAL

Biopulping

Wood was harvested and transported to a mill for debarking, chipping, and screening. Wood chips were decontaminated by steaming, maintaining a high temperature for a sufficient time to decontaminate the chip surfaces, and then cooled so that the fungus could be applied. Chips were then placed in piles that were ventilated to maintain the proper temperature and moisture content for fungal growth and subsequent biopulping. Retention time was from 1 to 4 weeks. Softened chips were then processed through conventional TMP processing. Specific details for the results reported in this paper were previously published (19).

Microwave Pulping

Based on previous studies, availability, and the common use as a species for TMP, black spruce (*Picea mariana*) logs were selected for microwave pretreatment and pulping. Logs were as “green” as possible (preferably about 40% moisture content). Logs were shipped to the pilot facility of MICRODRY, INC., a manufacturer of custom industrial microwave ovens.

A high capacity microwave oven was used. Various treatment levels were investigated based on power setting (20 to 50 kW) and exposure time (1 to 10 min). At the conclusion of the trials, all treated logs were individually shrink-wrapped and shipped to FPL for further processing.

The protocol incorporated an initial TMP fiberization pass to obtain a target freeness of 750 mL Canadian Standard Freeness (CSF), followed by four subsequent atmospheric refining passes to attain 550, 350, 200, and 100 mL CSF, respectively. Power consumption was measured for each refiner pass. Specific details of the results were previously published (9).

Bleaching and Fiber Loading of Bio- and Oxalic-Acid-Pretreated TMP

For producing oxalic acid pretreated TMP, southern yellow pine logs were debarked, chipped, and screened. Individual batches of 45.4 kg (ovendry (OD)) were submerged in about 0.3 m^3 of 1.0 % oxalic acid solution in a 0.33-m^3 stationary digester. The excess was then discharged. For the remaining chips, temperature was raised to 130°C and held at that temperature for 10 min. After treatment, chips were thoroughly washed with city water in a 100-mesh screen basket and stored at 5°C. After pretreatment, chips were shipped to Andritz, Inc. in Springfield, Ohio, for commercial-scale TMP preparation.

For combined fiber loading and bleaching of bio-pretreated TMP (bio-TMP), we used a mixture of aspen groundwood (GW) pulp, spruce TMP, and bleached mixed softwood kraft (SWBK) pulp in a typical furnish in a ratio of 22.5%, 27.5%, and 50%, respectively, to produce lightweight coated paper. The spruce TMP was the variable component of this furnish. TMP was produced from both control chips and bio-treated (inoculated with *Ceriporiopsis subvermispora* SS-3) chips at FPL. Kraft wetlap pulp was refined to 420 CSF using a 305-mm-diameter Sprout–Waldren (Springfield, OH) atmospheric refiner; TMP was prepared on a 305-mm-diameter Sprout–Bauer (Springfield, OH) pressurized refiner. Initial brightness levels (International Organization for Standardization (ISO)) of the separate pulps were as follows: GW, 62% to 64%; TMP control, 50% to 55%; bio-TMP, 40%; and SWBK, 80%. The brightness target for the blend was 72% to 74% ISO. It is possible to fiber load all types of fibers, but because the spruce TMP and GW portion of this furnish requires substantial bleaching to meet the brightness target, the mechanical pulp components GW and TMP were selected for simultaneous fiber loading and bleaching.

The dry calcium hydroxide used for fiber loading of bio-TMP was industrial-grade Mississippi Codex hydrated lime (Mississippi Lime Co, Alton, IL). Slurried calcium hydroxide was used for the oxalic acid TMP experiments. Comparative direct loading was done with papermaker-grade (HO) precipitated calcium carbonate (PCC) obtained from Specialty Minerals, Inc. (Bethlehem, PA).

For fiber loading, 1.5-kg batches of GW–TMP blends were prepared by adding dry calcium hydroxide (20% based on OD pulp) into moist pulp, followed by mixing thoroughly in a Hobart laboratory mixer (Troy, NY) at 20% consistency for 15 min. The pulp was added into the chamber of a Sprout–Bauer refiner and reacted with carbon dioxide at 207 kPa for 10 min retention before passing through the refiner at a wide plate (65-mm) gap. When peroxide was included, 2% H_2O_2 and typical stabilizers (3% sodium silicate, 0.05% magnesium sulfate) were mixed into the mechanical pulp blend at 20% consistency prior to adding calcium hydroxide into the Hobart mixer. The pulp was reacted with CO_2 as previously described and passed through the refiner. Exit temperature of the pulp was approximately 40°C to 45°C. Fiber-loaded pulp (containing peroxide) was transferred into a plastic bag and held for 1 h at 60°C. After bleaching, the fiber-loaded pulp was blended 50:50 with SWBK for handsheet preparation and testing following TAPPI methods.

Handsheets (60 g/m^3) for direct loading were made (TAPPI T-205) by first mixing equal portions of GW–TMP blend with SWBK. Papermaker's PCC was then added directly into the doler tank to the approximate ash level of the fiber-loaded handsheets. Considerable excess of PCC was required to retain a target ash level in handsheets. The dual polymer retention aids for experiments containing oxalic acid pretreated TMP were cationic (polyacrylamide, PL 2310) and anionic (colloidal silica, NP 890) materials obtained from Eka Chemicals, Inc. (Marietta, GA). Handsheets were made and evaluated for physical and optical properties according to TAPPI standard test method T-220. Ash content was measured at 400°C according to T211.

For fiber loading of oxalic acid pretreated TMP, treated pulps were prepared into handsheets without blending with other pulps.

Table I. Energy savings and paper properties for various pretreatments for TMP at 100 mL CSF

Pulping method	Energy savings (%)	Tensile index (Nm/g)	Tear index ($mN\text{-}m^2/g$)	Brightness (% ISO)	Scattering coefficient (m^2/g)
Bio-treated spruce chips (bio-TMP)					
Control	—	44.6	9.14	53.1	55.8
Treated[a]	31	47.5	8.33	31.0	36.7
Microwaved spruce logs					
Control	—	27.4	4.07	56.0	51.7
Treated[b]	10	31.0	4.88	50.7	49.3
Oxalic acid treated southern yellow pine chips					
Control	—	30.2	7.4	52.8	53.4
Treated[c]	20	29.6	8.4	51.7	51.9

[a]Pretreatment at FPL. Mechanical pulping at Andritz, Inc. (Springfield, OH) using 91-cm pressurized refiner in stage 1 and 91-cm refiner in subsequent stages.
[b]Pretreatment at Microdry, Inc. (Louisville, KY). Mechanical pulping at FPL using 30-cm pressurized refiner in stage 1 and 30-cm refiner in subsequent stages.
[c]Pretreatment at FPL. Mechanical pulping at Andritz, Inc. using 91-cm pressurized refiner in stage 1 and 91-cm refiner in subsequent stages.

RESULTS

Energy savings are possible by any of the three pretreatments investigated. The largest energy saving we report is 31% for the bio-TMP (Table I). Microwave pretreatment reduced energy consumption by 10% and oxalic acid pretreatment by 20%. The bio-TMP results reported were from many developmental studies and represent an accurate prediction of the energy reduction that can be expected of a commercial application. The microwave and oxalic acid pretreatment results were from preliminary experiments and need optimization. For example, the oxalic acid pretreatment has not been optimized regarding chip penetration of the acid solution, and this process should only stand to benefit with more complete chip penetration.

TMP from microwave pretreated logs was produced using a 30-cm diameter refiner, and the experiments need to be developed for production on larger industrial-scale refiners. The microwaved TMP shows promise since its brightness was reduced only 5.3 percentage points compared to 22.1 percentage points for the bio-TMP. Brightness loss is indicative of chemical change induced by either the biopulping reactions or the rapid heating of the logs by microwaving.

Handsheets prepared from oxalic acid pretreated TMP showed an even smaller reduction in brightness of only 1.1 percentage points. In addition, the scattering coefficient loss was 1.5 percentage points for oxalic acid TMP compared with 19.1 percentage points for bio-TMP. Both microwave and oxalic acid pretreatments resulted in greatly improved optical properties compared to that of bio-TMP, which was the least effective with respect to the preservation of optical properties. Bio-pretreatment resulted in tensile and tear index changes of 2.9 and −0.81 points, respectively; microwave pretreatment 3.6 and 0.81, respectively; and oxalic acid pretreatments −0.6 and 1.0 points, respectively.

Microwave pretreatment resulted in the greatest overall improvement in handsheet strength properties, and bio-pretreatment resulted in the greatest improvement in energy reduction. All three pretreatments reduced brightness and scattering coefficient.

Adding inorganic filler is the conventional method for improving brightness and scattering coefficient of paper. Using bio-TMP as part of the furnish and combining it with groundwood pulp before fiber loading the two mechanical pulps, then combining the furnish with 50% SWBK, resulted in an improvement in optical properties compared to that of identical handsheets made with standard TMP. That is, fiber loading made a greater improvement in brightness and scattering coefficient when processing a furnish containing bio-TMP

Table II. Effect of fiber loading on TMP and bio-TMP after blending with 50% SWBK and increasing mechanical/SWBK ratio[a]

Handsheet samples	Tensile index (Nm/g)	Tear index ($mN\text{-}m^2/g$)	Brightness ISO (%)	Scattering coefficient (m^2/kg)	$CaCO_3$ (%)
Control, 50/50	21.1	9.95	67	60	9.4
Treatment, 50/50	23.9	11.07	67	64	8.7
Treatment, 55/45	23.7	10.52	68	72	9.1
Treatment, 60/40	21.5	9.90	68	79	11.0

[a] TMP is thermomechanical pulp; GW, groundwood; SWBK, bleached mixed softwood kraft.

than did fiber loading a similar furnish containing standard TMP. Table II shows an improvement in scattering coefficient from 60 m^2/kg for the control to 64 m^2/kg for the fiber-loaded bio-TMP furnish. Brightness remained the same (67%) for both experiments. However, ash content was 8.7% for bio-TMP as opposed to 9.4% ash for control TMP. The lower ash content of bio-TMP would tend to lower brightness. Since brightness remained the same for both pulps, even greater brightness might be achievable with bio-TMP at higher ash content. Since strength properties were higher for bio-TMP than for the control, the possibility of replacing fiber with filler exists. This would be a favorable replacement economically.

The strength advantage of fiber loading over direct loading permitted the ratio of mechanical pulp to softwood pulp to be increased from a 50:50 to 60:40. The reduction of 10 percentage points for bleached softwood kraft and replacement with mechanical pulp represents an obvious saving in fiber cost.

Preliminary experiments for fiber loading of oxalic acid pretreated TMP compared with standard TMP resulted in an initial strength advantage for the oxalic acid pretreated TMP. Table III shows tensile index results for fiber-loaded oxalic acid pre-treated TMP compared with fiber-loaded standard TMP (control). The strength of the oxalic acid pretreated TMP was higher up to about the 18% ash level, albeit at a somewhat lower differential than at the 0% level.

In another preliminary experiment, we combined peroxide bleaching with fiber loading and direct loading of oxalic acid treated TMP and compared the results to standard TMP. Table IV shows the resulting tensile indexes and filler levels. Fiber loading and conventional direct loading resulted in similar tensile indexes under the conditions used. Again, a strength differential was maintained between the oxalic acid pretreated TMP and the standard TMP for both fiber loading and direct loading.

Table III. Effect of fiber loading on TMP and oxalic acid pretreated TMP

Handsheet sample	$CaCO_3$ (%)	Tensile index (Nm/g)
Control	0	14.91
	5.5	12.98
	16.1	10.36
Oxalic acid pretreated chips	0	18.49
	5.8	15.14
	18.8	10.52

Table IV. Effect of fiber loading with H_2O_2 versus direct loading on TMP and oxalic acid pretreated TMPs

Handsheet sample	Filler method	Tensile index (Nm/g)	$CaCO_3$ (%)
Control	Fiber loaded	10.09	9.3
	Direct loaded	9.26	8.4
Oxalic acid treated	Fiber loaded	11.87	10.9
	Direct loaded	12.24	10.6

CONCLUDING REMARKS

For the conditions reported here, TMP produced from various species and pretreatments at 100 mL CSF showed differences in energy requirements for pulp production, handsheet properties prepared from pulps, and response of pulps to PCC added conventionally and calcium carbonate added by fiber loading.

The greatest reduction in energy occurred for bio-TMP (31%). Microwave and oxalic acid treatments resulted in a reduction in energy of 10% and 20%, respectively. Handsheet strength properties were improved to about the same extent for all three pretreatments. Optical properties of brightness and scattering coefficient were reduced the least with oxalic acid and microwave treatments compared with bio-pretreatment.

Fiber loading of bio-TMP improved handsheet strength compared to that of similar untreated TMP to the extent that strength improvement permitted a reduction in 10 percentage points in bleached softwood kraft reinforcing pulp. Fiber loading of TMP produced from oxalic acid pretreated chips compared to that of control TMP maintained the strength advantage of the oxalic acid pretreated TMP. Fiber loading and direct loading of oxalic acid pretreated pulps resulted in similar handsheet strengths.

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