ORGANOSOLV PULPING OF POPLAR WOOD FROM SHORT-ROTATION INTENSIVE CULTURE PLANTATIONS

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ABSTRACT

This study investigates the suitability of the ethanol organosolv process to pulp poplar wood from short-rotation intensive culture (SRIC) plantations. Wood chips were prepared from 9-year-old SRIC debarked poplar logs (*Populus deltoides* × *trichocarpa* '*Donk*'), and they were subjected to ethanol/water organosolv pulping in lab-scale batch reactors. The effect of the main process variables (i.e., time, temperature, ethanol concentration) on the pulp yield and quality was examined. Upon process optimization, the papermaking properties of the pulp obtained were determined.

The most selective conditions in terms of delignification and pulp viscosity were found at an ethanol concentration of 60% (v/v) in water, a pulping time of 210 min and, a temperature of 195 C. Under these conditions, good delignification was achieved (kappa no. 20.2), with pulp yields of 54.3%, and degree of polymerization (DP) values ranging from 1,900 to 2,000. The strength properties of hand-sheets made from the SRIC poplar organosolv pulp were in the same range, except for a lower tear factor, as those reported for hardwood (semi-) chemical pulps. The results obtained indicate the promising potentials of the organosolv process for the chemical pulping of fast-grown poplar wood cultivated in short rotations.

Keywords: *Populus,* short-rotation intensive culture poplar hybrids, juvenile wood, organosolv pulp, yield, kappa, degree of polymerization, handsheet properties.

INTRODUCTION

The rapid and steady increase in worldwide demand for timber and forest products, coupled with the declining availability of wood supplies, is placing increasing emphasis on the potentials of wood from short-rotation intensive culture (SRIC) plantations as a raw material in the pulp and paper industry. Poplar hybrid clones, which have a high growth rate, offer big promise as raw materials for pulp and papermaking. Previous research has shown that the wood obtained from several fast-growing hybrid poplar species cultivated in rotations of only 2 to 12 years is suitable for the production of hardwood chemical and semi-chemical pulps (Dix and Roffael 1989; Hunt and Keays 1973; Labosky et al. 1983; Marton et al. 1968; Snook et al. 1986). Phelps et al. (1985) also demonstrated the potentials of SRIC poplar wood for supplementing conventional conifer furnishes for kraft pulping. In these studies, the strength properties of SRIC poplar pulps were generally found to be similar or only slightly lower than those of pulps made from mature poplar chips pulped under the same conditions.

Solvent pulping technologies have received

significant attention in recent years as an attractive alternative to conventional chemical processes. Organosolv pulping offers several economic and environmental advantages over the established chemical pulping methods, such as low chemical and energy consumption, simple solvent and by-product recovery, suitability for small-scale chemical pulping operations, relatively low capital investment required for a new mill and, low environmental impact (Paszner and Cho 1989). Organosolv pulping has been shown to offer good prospects for the manufacture of hardwood chemical pulps and could be a viable alternative for the manufacturing of SRIC poplar chemical pulps.

Organosolv pulping is a sulfur-free chemical pulping process in which aqueous organic solvents are used to produce bleachable-grade pulps. Solvent pulping systems evaluated at the pilot-scale or already at full-industrial scale utilize aqueous mixtures of ethanol (Pye and Lora 1991), methanol (Edel 1989), or acetic acid (Nimz et al. 1986). Organosolv pulping has been shown to be particularly suitable for the delignification of hardwoods (Aziz and Sarkanen 1989) and annual fibers (Tjeerdsma et al. 1994; Winner et al. 1991). However, some variants of the process are applicable to both hardwoods and softwoods. The addition of catalysts such as mineral acids, organic acids, or alkali earth metal salts, among others, appears to be necessary to pulp softwoods (Paszner and Cho 1989). Results obtained with some solvent processes are very promising, with high pulp yields at strength values that match those of sulfite and even kraft pulps from the same wood species (Aziz and Sarkanen 1989; Lora and Aziz 1985). In addition, organosolv pulps can easily be bleached using chlorine-free bleaching sequences (Cronlund and Powers 1992).

The objective of this research was to investigate the suitability of the ethanol organosolv process to pulp SRIC poplar (9-year-old *Populus deltoides* × *trichocarpa* '*Donk*') wood. Organosolv pulping was optimized based on the quantity and properties of the pulp produced under different cooking conditions. Further-more, the papermaking properties of the pulp obtained upon process optimization were tested and compared with those of commercial semi-chemical and chemical pulps.

MATERIALS AND METHODS

Raw material

Three 9-year-old *Populus deltoides* × *trichocarpa* '*Donk*' trees were harvested at random from a poplar stand cultivated on former grass farmland located within the municipality of Ede, The Netherlands. The average height of the trees was 14.3 m and their average diameter at breast height 0.20 m. The stems (over the total tree height) were harvested during the dormant season after leaf fall. The debarked stems were chipped and subsequently combined to form a composite sample. The chips were dried at 30 C in a chamber with forced dry-air circulation to a moisture content of approximately 5% and then screened using a 10-mm screen. Oversized chips and fines were discarded.

Pulping

Pulping experiments were carried out according to methods described in earlier work (Tjeerdsma et al. 1994; Tjeerdsma and Zomers 1993). Wood chips were pulped in ethanol/water mixtures, without catalyzing additives. Prior to pulping, the chips were impregnated with cooking liquor by boiling for 15 min under reflux. Pulping was performed in oil-heated rocking batch reactors. Batch reactors of 0.1 liter and 4 liter volume were used to study the effect of pulping conditions and to produce enough pulp for papermaking, respectively. Time, temperature, and ethanol concentration were varied in order to find optimum pulping conditions. The liquor to wood (l/w) ratio was kept constant in all experiments at 8 ml/g. Following pulping, the reactors were cooled in a water bath, and samples of the cooking liquor were withdrawn. All pulps were disintegrated in a laboratory blender, washed three times with a hot ethanol/water (1:1, v/v) mixture, and finally washed with hot deionized water.
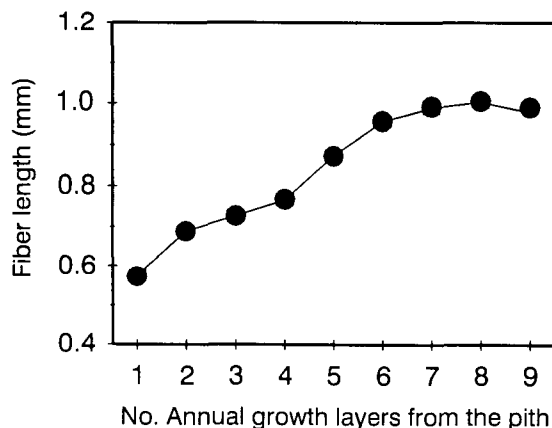
FIG. 1. Radial variation of the fiber length in the wood from 9-year-old *Populus deltoides* × *trichocarpa* 'Donk' trees.
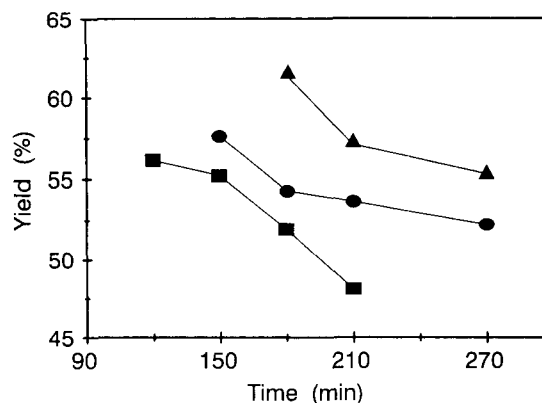

FIG. 2. Effect of the cooking time on the yield of SRIC poplar pulp when pulping with 60% (v/v) ethanol in water at a temperature of 185 (▲), 195 (●) and, 205 C (■).

Chemical analysis

Pulp yield (expressed as percentage of the oven-dry weight of the chips) was determined gravimetrically after drying 24 hours at 60 C. Kappa number measurements were performed according to TAPPI testing procedure T 236 cm-85. Pulp viscosity was determined according to a modified TAPPI method T 230 om-82 and expressed as degree of polymerization (DP) of cellulose. The klason lignin and acid-soluble lignin content of the wood were determined according to TAPPI standards T 222 om-83 and um-250, respectively. The holocellulose content was determined according to the acid chlorite method described by Browning (1967). Wood extractives were determined according to a modified TAPPI method T 204 om-88 using ethanol-cyclohexane (1:2, v/v) as organic solvent.

The average chemical composition of the wood chips used in this study, expressed as percentage of the wood dry weight, was: lignin 18.6%, holocellulose 71.5%, and extractives 1.3%.

Fiber length analysis

The fiber length and width were measured in stem cross sections taken at relative heights of 25, 50, and 75% of the total tree height. Wood samples of 3 × 3 × 20 mm were taken from each annual ring and macerated in a 1:2 (v/v) mixture of acetic acid and hydrogen peroxide (30%) at 100 C for 3 hours. The fiber dimensions were measured using an overhead light transmission microscope.

The mean weighted average fiber length was 0.886 mm. The fiber length to diameter ratio averaged 34.53 μm/μm. Figure 1 shows the radial variation in the fiber length. An increase in fiber length with increasing distance from the pith is evident from these results.

Handsheet preparation and testing

The fiber pulp was beaten in a PFI mill according to TAPPI method T 248 cm-85. Handsheets were formed and tested for burst, breaking length, and tear according to modified TAPPI methods T 205 om-81 and T 220 om-83.

RESULTS AND DISCUSSION

The ethanol organosolv pulping of short-rotation intensive culture (SRIC) poplar wood was optimized. The effect of three pulping conditions, i.e., time, temperature, and ethanol concentration, on the pulp yield, lignin content (kappa number), and viscosity (DP) was investigated.

Figures 2 and 3 illustrate the effect of cooking time on the extent of delignification and pulp yield when the SRIC poplar wood was
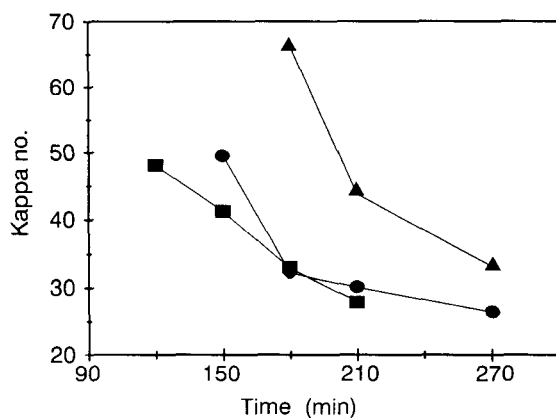
FIG. 3. Effect of the cooking temperature on SRIC wood delignification (expressed as kappa no.) when pulping with 60% (v/v) ethanol in water at a temperature of 185 (▲), 195 (●) and, 205 C (■).

FIG. 4. Effect of the ethanol concentration on SRIC wood delignification (temperature = 195 C; time = 210 min).

pulped at various temperatures in a liquor containing 60% (v/v) ethanol. As shown in Fig. 2, delignification was greatly enhanced with increasing reaction time and pulping temperature. Increasing the pulping temperature from 185 to 195 C reduced residual lignin by 50% or more depending on cooking time. A further increase of the temperature from 195 to 205 C only caused a minor decrease in the kappa number. Pulping at 205 C resulted in a steady decrease of the kappa number with cooking time during the period investigated. In contrast, delignification at lower temperatures proceeded through two kinetically distinct phases. A relatively rapid decrease in the kappa number was observed in the initial period, followed by a much slower delignification rate in the later stage of the cook. Previous kinetic studies have shown that during the early stages of organosolv pulping, a rapid and preferential delignification of syringyl lignin occurs and is followed by a slower removal of guaiacyl lignin (Goyal et al. 1992).

The effect of cooking time and temperature on the pulp yield (Fig. 3) followed the same general trends described above for the delignification. However, increasing the pulping temperature from 195 to 205 C resulted in a considerable decrease in the pulp yield. The
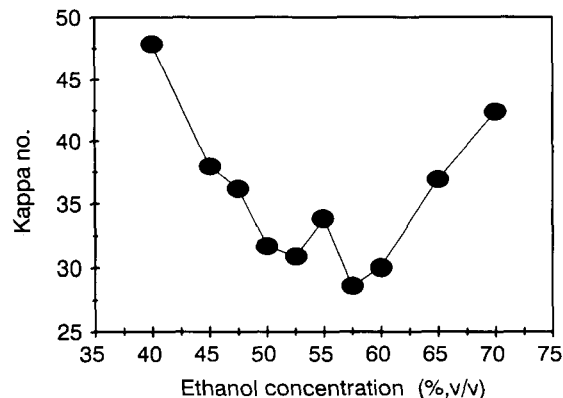
pulp yield loss was not accompanied by a decrease in the kappa values, suggesting that the increased material losses at 205 C were mainly due to polysaccharide degradation. Based on these results, a cooking temperature of 195 C appears to be desirable in order to maximize both delignification and pulp yield. Under these conditions, a pulping time of 210 min was sufficient for suitable delignification.

The ethanol concentration was found to have a major impact on the quality and quantity of the pulp (Fig. 4). Optimal delignification, as evidenced by the lowest kappa, was obtained at alcohol concentrations ranging from 50 to 60% (v/v). Pulping at higher ethanol concentrations resulted in an almost linear increase of the residual lignin. This trend can be attributed to the decreasing acidity of the cooking liquor with increasing ethanol concentrations (Fig. 5). Previous studies have reported that acidity plays a crucial role in solvent pulping, because lignin dissolution results from the acid catalyzed cleavage of α-aryl and β-aryl ether linkages in the lignin macromolecule (Goyal et al. 1992; Lai and Guo 1992). Poor pulp delignification was also obtained with diluted alcohol solutions, due to the lower dissolution of lignin and the enhancement of lignin condensation reactions under the high acidity conditions prevailing (Goyal and Lora 1991; Paszner and Cho 1989).
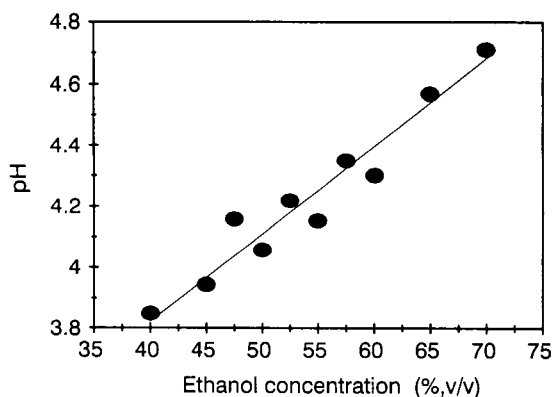
FIG. 5. Effect of ethanol concentration on the pH of the cooking liquor after organosolv pulping of SRIC poplar wood (temperature = 195 c; time = 210 min).
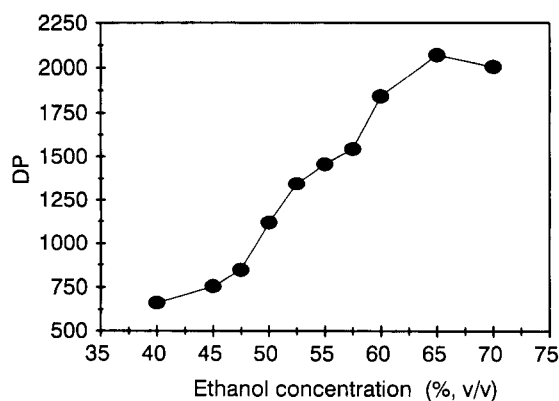

FIG. 7. Effect of the ethanol concentration on the viscosity of SRIC poplar pulp (temperature = 195 C; time = 210 min).

Similarly, the ethanol concentration applied during pulping was found to have a major effect on both the pulp yield and the pulp viscosity. A positive relationship was observed between the alcohol content in the pulping liquor and these pulp parameters (Figs. 6 and 7). The decrease in pulp yield with decreasing ethanol concentration is the result of the more extensive hemicellulose hydrolysis and dissolution occurring under conditions of increasing acidity. Cellulose, on the other hand, is largely conserved in autocatalyzed organosolv pulps when diluted ethanol mixtures are used. However, some losses in pulp viscosity do occur, which can be attributed to partial depo-
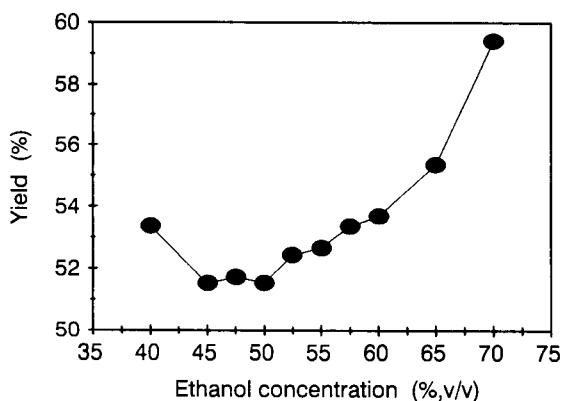
lymerization of cellulose by acid-catalyzed hydrolysis (Goyal et al. 1992).

In the range of pulping conditions investigated, it appears that an ethanol concentration of 60% (v/v), a pulping time of 210 min and, a temperature of 195 C provide a good compromise. An adequate rate of delignification to a kappa no. of 30.1 is achieved while retaining a reasonable pulp viscosity of 1,845 DP. Under these conditions, a high pulp yield of 53.7% was also obtained. These pulp parameters are within the range of those obtained in experimental kraft pulping of poplar (Hunt and Keays 1973; Labosky et al. 1983; Snook et al. 1986).

In order to obtain sufficient quantities of pulp for paper testing, additional pulping experiments were conducted in a 4 liter digester. The yield and DP values of the pulp produced were similar to those of the pulp originating from the 0.1 liter digester (Table 1). However, the pulp from the larger reactor had a lower kappa number of 20.3. The improved delig-

FIG. 6. Effect of the ethanol concentration on the yield of SRIC poplar pulp (temperature = 195 C; time = 210 min).

TABLE 1. *Results obtained during the organosolv pulping of SRIC poplar wood in a 4 1 batch reactor (ethanol concentration = 60% (v/v) in water, temperature = 195 C, time = 210 min).*

Yield (%)	54.3
Kappa no.	20.2
Degree of polymerization	1,959

TABLE 2. *Comparison of the mechanical properties of handsheets made from SRIC poplar organosolv pulp with those of hardwood semi-chemical and chemical pulps.*

Pulp	Breaking length (km)	Tear factor	Burst factor
SRIC poplar organosolv	6.8	39.0	36.9
Hardwood organosolv	6.4–10.2	64.5–78.4	49.6–66.5
Poplar kraft	5.4–9.0	65.0–79.0	38.0–52.1
Hardwood kraft	4.5–11.6	45.9–98.9	31.0–57.0
Poplar sulfite	5.7–8.6	—	22.4–58.1
Hardwood sulfite	7.2–8.7	28.9–107.0	35.0–50.7
Poplar NSSC	5.0–7.8	—	21.5–52.1

References. Dix and Roffael 1992; Dix et al. 1992; Hunt and Keays 1973; Lora and Aziz 1985; Pye and Lora 1991; Roffael and Dix 1994; Snook et al. 1986.

nification can be explained by a different pulp washing procedure employed in the latter case, which utilized 0.5% NaOH instead of hot aqueous alcohol solutions. Evidently, the dilute NaOH solution was more effective in removing lignin adsorbed on the pulp fibers. Condensation and partial reprecipitation of the solubilized lignin are known to occur during the late stages of organosolv pulping and upon cooling of the pulping liquor (McDonough 1993; Tirtowidjojo et al. 1988). The condensed lignin-fraction is for the most part insoluble in aqueous alcohol solutions (Paszner and Cho 1989).

The strength properties of handsheets made from the organosolv pulp are shown in Table 2. Literature data of handsheets from chemical and semi-chemical pulps of poplar and other hardwoods are also listed for comparison. The tensile and burst strength properties of the SRIC poplar organosolv pulps were within the same range as those of hardwood NSSC, sulfite, and kraft pulps, or bleached organosolv pulps from mixed North American hardwoods. In contrast, the tear strength of the SRIC poplar organosolv pulp was poor. The low tear values can be attributed to the short fiber length of 0.886 mm in the juvenile poplar wood.

The poplar organosolv handsheets showed a relatively low brightness of 23.2% ISO, as compared to chemical pulps from the same wood species. Brightness levels reported for unbleached kraft poplar pulps, ranged from 28.0 to 40.2% ISO (Dix et al. 1992). The poor

optical properties of the organosolv pulp should not be considered as a critical obstacle for the application of this process, because organosolv pulps are known to be highly receptive to bleaching (Cronlund and Powers 1992).

CONCLUSIONS

This research shows that SRIC poplar wood can be pulped to adequate degree of delignification (kappa no. 20.2) and DP values (1,900–2,000), with an excellent yield (54.3%) by means of the organosolv pulping process. The most selective delignification conditions were found at an ethanol concentration of 60% (v/v), a pulping time of 210 min, and a temperature of 195 C. Organosolv pulp from the 9-year-old poplar trees had burst and tensile strength properties comparable to those of chemical and semi-chemical hardwood pulps. However, lower tear strengths were determined due to the low fiber length in the juvenile wood pulped. These results suggest that organosolv pulping can be a viable alternative for the production of chemical pulps using fast-grown poplar cultivated in short rotations.

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