Pulp Extrusion: A New Processing Method for Recycling Recovered Wastepaper and Papermill Sludge and its Application for Building Materials

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Abstract

Fibers recovered from recycled paper products have desirable mechanical properties and can be used for the manufacture of composite materials. Contrary to their use in papermaking, recycled wood pulp fibers used for composites may require only minimal cleaning and de-inking, thus greatly reducing manufacturing costs. In addition, waste fiber sources which are not used presently could be exploited.

In the research presented it is shown that extrusion processing of highly concentrated, aqueous pulp fiber suspensions offers a way to manufacture composite materials which make use of the inherent strength and bonding potential of wood fibers. With this process, fiber composites with mechanical properties comparable to those found in medium density fiberboard and hardboard can be produced continuously. Stiffness loss due to moisture can be controlled by addition of crosslinkable resins, thus offering the possibility to manufacture structural materials with consistent and predictable properties. Tolerance of the extrusion process toward the addition of inorganic materials make it a suitable process for recycling contaminated fibers and papermill sludges.

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Introduction

Fibers recovered from recycled waste paper have desirable mechanical properties and are already widely used in the manufacture of paper and paperboard. Wood fibers have anisotropic strength and stiffness properties; both are higher along the fiber length axis and are a function of fiber type, fibril angle, processing history, and defects (7,8). Tailored mechanical properties of composites can thus be achieved by controlling fiber orientation during the formation process.

Paper and paper products constitute about 40 percent (71 million tons/yr.) of municipal solid waste in the United States. At present, recyclability of this waste material is predicated on successful collection, cleaning, and de-inking, yielding fibers that can be used in conventional paper and board making (1). Although the supply of waste paper is vast, this resource is not yet sufficiently used because the costs associated

with separating fillers and contaminants from fibers can be considerable and fiber yields are low.

Another persistent problem in the paper industry, entailing both economic and environmental costs, is the disposal of papermaking sludges. Sludge is the generic term for the solid residue, containing inorganic fillers and fiber fines, recovered from the wastewater stream of pulp and papermaking operations as well as recycling operations. The paper industry in the United States produces an estimated 4.1 million tons of sludge each year, of which 69 percent are landfilled and 29 percent are incinerated(9). Processing technology on industrial scales to recycle this waste material is lacking. New processes that convert contaminated waste fibers and sludges into useful products are therefore necessary.

High consistency forming methods that do not depend on suspension drainage on a screen are desirable because they make use of the inherent strength and bonding potential of wood fibers. Their implementation needs to overcome two problems. First, high-consistency pulp suspensions must be made to flow homogeneously to achieve good formation. Second, processing equipment which can disperse fiber floes and transport highly concentrated pulp suspensions must be found.

Pulp suspensions at high consistencies are usually severely flocculated and thus do not flow homogeneously at low shear rates. This problem can be overcome by adding water-soluble polymers to concentrated pulp suspensions (4–6, 12, 13) and hydrophilic papermill sludges (14). After polymer addition, these suspensions flow easily and homogeneously at consistencies up to 45 percent. The water-soluble polymers bind free water in the suspension and apparently reduce interfiber friction as well as friction between fibers and processing equipment. Most importantly the rheology of pulp suspensions can be controlled such that the suspensions can be processed with extrusion equipment typically used in the food and plastics processing industries.

Process technology

A high-consistency pulp extrusion process was recently developed at the USDA Forest Products Laboratory (Fig. 1). Dewatered crumb pulp (typically 30% consistency) is blended with a high molecular weight, water soluble polymer (typically 3% , based on dry weight fiber) and optional additives. This blend is fed into an extruder where it is intensively mixed, dispersed, and extruded through a die. The die determines the cross-sectional shape of the extrudate and fiber alignment. The extrudate is then dried and, if desired, densified. The final product has a moisture content of typically 8 percent and can contain over 90 percent fibers.

High consistency processing by twin-screw extrusion

It was found that twin-screw extruders are well suited to mix, disperse, and transport highly concentrated pulp fiber suspensions. Compared with single-screw extruders, twin-screw extruders have several advantages. First, they act as positive displacement pumps. Second, they have a flexible screw design to tailor mixing and dispersion action and, finally, they can be used as chemical reactors.

A key to successful pulp extrusion is an understanding of mixing and dispersion processes because the dispersion of fiber floes is difficult at high consistencies. Dispersive mixing occurs predominantly in regions of the twin-screw extruder where shearing actions are present and can be enhanced by employing kneading elements. The mixing and dispersion action can be tailored to accommodate different pulp types and additives by judicious arrangement of transport and kneading elements on the screws.

Experimental

The results from several investigations are summarized and discussed. A laboratory-scale, co-rotating twin-screw extruder (Leistritz Micro, 18 mm) was used in all experiments. Three fiber types were used (Table 1). Extruded material was dried and densified in a laboratory press (temperature 150°C; pressure 350 kPa; press time 3 min). The dried extrudate was conditioned at 50 percent relative humidity before tension tests were performed. The modulus of elasticity (MOE) was determined by a first order linear regression fit to stress-strain data points below 0.1 percent strain. Table 2 shows the suspension compositions and extrusion conditions for all experiments.

Results and discussion

To determine suitable extrusion conditions, the dependence of pressure, screw torque, and residence time on screw speed was studied. It was found that screw torque and residence time decreased nonlinearly with increasing screw speed (i.e., increasing shear rate) whereas die entry pressure remained constant (Fig. 2). This suggests a shear thinning behavior of the concentrated pulp fiber suspension. As a consequence, in practice, an optimal operating point must be found, where the gain of increased mass flow at

high screw speeds is offset by the increased energy consumption of the extruder.

To exploit the mechanical properties of recycled fibers, fiber damage during high consistency processing must be kept to a minimum. The fiber length distribution in the extrudate after four passes was compared with that of the stock suspension to assess the extent of fiber shortening which occurred in recycled newsprint pulp due to mechanical working in the extruder. It was found that the fines fraction (1.00 mm) increased at the cost of medium and long fiber length fractions. Despite the severe treatment, only an equivalent of 11.5 percent of the fibers were shortened (Fig. 3).

One advantage of high consistency forming is that pulps with high filler content and papermill sludges can be processed without intensive washing and cleaning, since for formation suspension drainage on a wire is not required. The extrusion behavior and the mechanical properties of the extrudate will, however, depend on the type and volume fraction of filler in the pulp. In the present research it was found that fillers reduced both strength and stiffness (Figs. 4 and 5) of

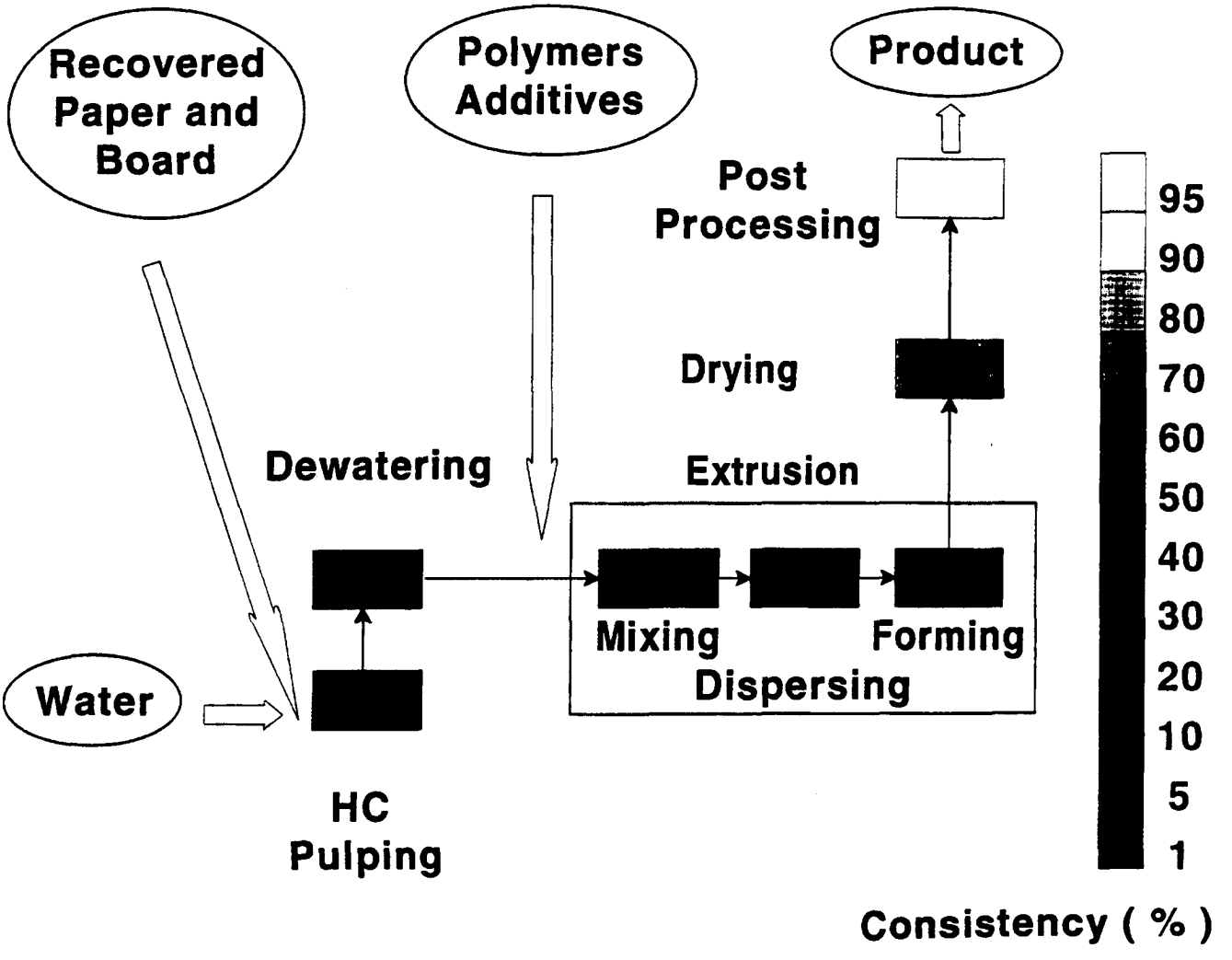

Figure 1.—Extrusion process schematic.

Table 1.—Pulp and fiber characteristics of pulps used in extrusion experiments.

Type	Species	Pulp type	Average fiber length	Ash content
			(mm)	(%)
Recycled, unprinted newsprint (ONP1)	*Picea* spp.	Thermomechanical	1.63	<2
Recycled, unprinted newsprint (ONP1)	*Picea* spp.	Thermomechanical	1.56	<2
Recycled *Smithsonian* magazine (MAG)	--	--	--	~26

Table 2.—Summary of experimental runs, pulp types, addition levels, and extrusion conditions.

Run no.	Fiber type[a]	Consistency	Polymer[b]	Additive		Screw speed	Torque	Die pressure	Temperature	Residence time
				Type[c]						
		(%)	(%)		(%)	(rpm)	(%)	(MPa)	(°C)	(sec.)
1a	ONP1	31	3	--	--	60	28	2.3	70	115
1b	ONP1	31	3	--	--	90	25	2.4	70	--
1c	ONP1	31	3	--	--	120	20	2.3	70	67
1d	ONP1	31	3	--	--	150	20	2.2	70	65
1e	ONP1	31	3	--	--	250	20	2.3	70	34
1f	ONP1	31	3	--	--	350	20	2.6	70	25
2	ONP1 control	31	3	--	--	140	23	4.3	70	--
3a	ONP1	31	3	sludge	10	190	22	3.5	70	--
3b	ONP1	31	3	sludge	20	190	22	2.8	70	53
4a	MAG	41	3	--	--	180	21	1.8	70	59
4b	MAG	41	1.5	--	--	180	22	2.5	70	54
4c	MAG	31	3	--	--	180	17	0.6	70	--
5	ONP2 control	31	3	--	--	180	20	2.2	70	--
6a	ONP2	32	3	PF1	5	180	20	1.9	70	--
6b	ONP2	32	3	PF1	10	180	19	1.9	70	--
6c	ONP2	35	3	PF1	20	180	18	2.1	70	--
7a	ONP2	32	3	PF2	5	180	18	1.9	70	--
7b	ONP2	32	3	PF2	10	180	15	1.5	70	--
7c	ONP2	35	3	PF2	20	180	--	1.7	70	--
8a	ONP2	32	3	MF	5	180	--	2.1	70	--
8b	ONP2	32	3	MF	9	180	20	--	70	65
8c	ONP2	35	3	MF	18	180	19	1.7	70	55
9a	ONP1	31	3	PAE	0.5	180	13	3.0	70	53
9b	ONP1	31	3	PAE	1.0	180	12	2.8	70	48
9c	ONP1	31	3	PAE	2.0	180	13	3.0	70	50

[a] ONP1, ONP2 = recycled, unprinted newspaper; MAG = recycled magazines. See Table 1 for characteristics.

[b] Sodium carboxymethyl-cellulose (Aqualon, type 7H4-F).

[c] sludge = aluminasilicate and cellulose fiber, ~49 percent ash; PF1 = phenol-formaldehyde resin (Plenco, type 11782); PF1 = phenol-formaldehyde resin (Plenco, type 12473); MF = melamine-formaldehyde resin (American Cyanamid Co., type Parez 707); PAE = Cationic poly(amino)amide-epichlorohydrin (Hercules, type Kymene 557H).

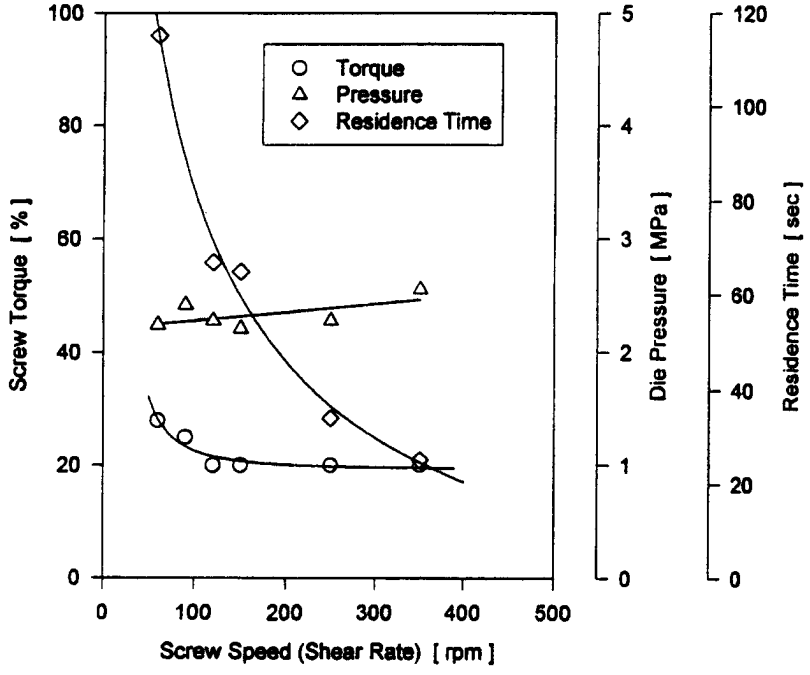

Figure 2.—Dependence of screw torque, die pressure, and residence time on screw speed (runs 1a-f).

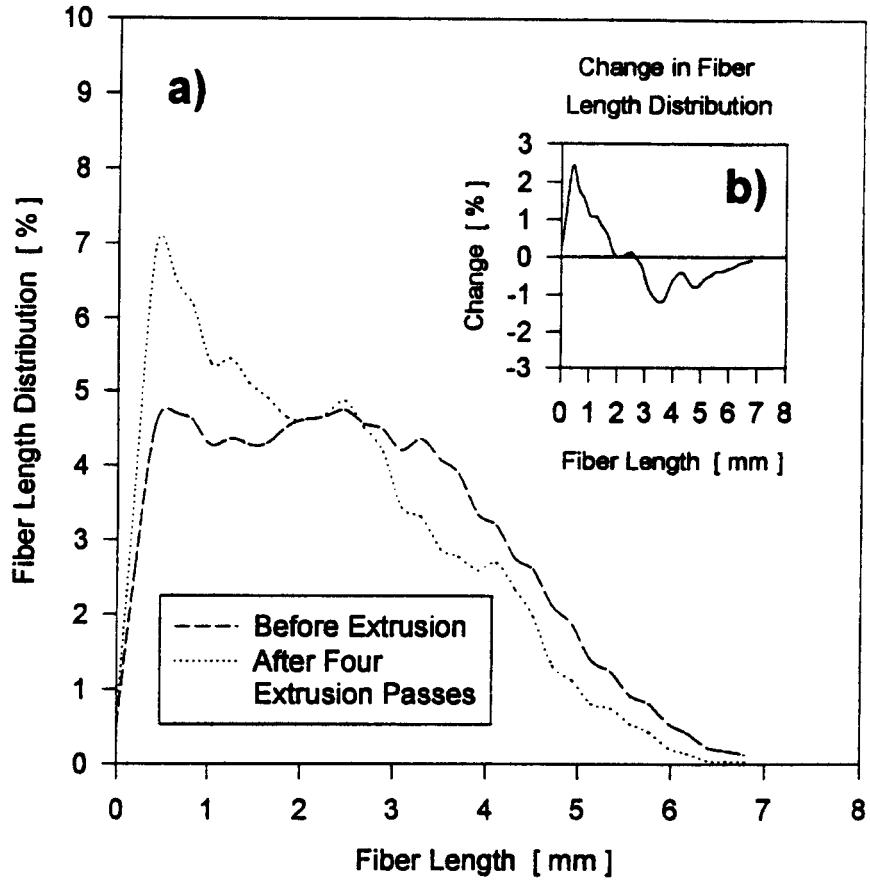

Figure 3.—Effect of shear history on (a) fiber length; and (b) change in fiber length direction after four extrusion passes.

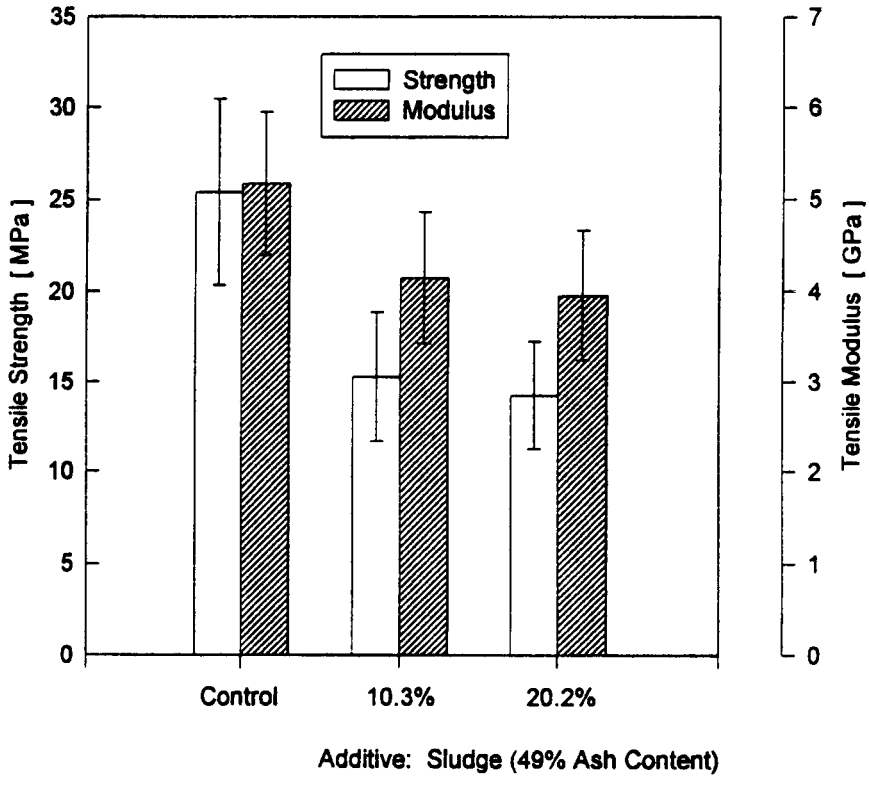

Figure 4.—Tensile strength and tensile modulus for extrudate containing 10 percent (run 3a) and 20 percent (run 3b) papermill sludge (49% ash).

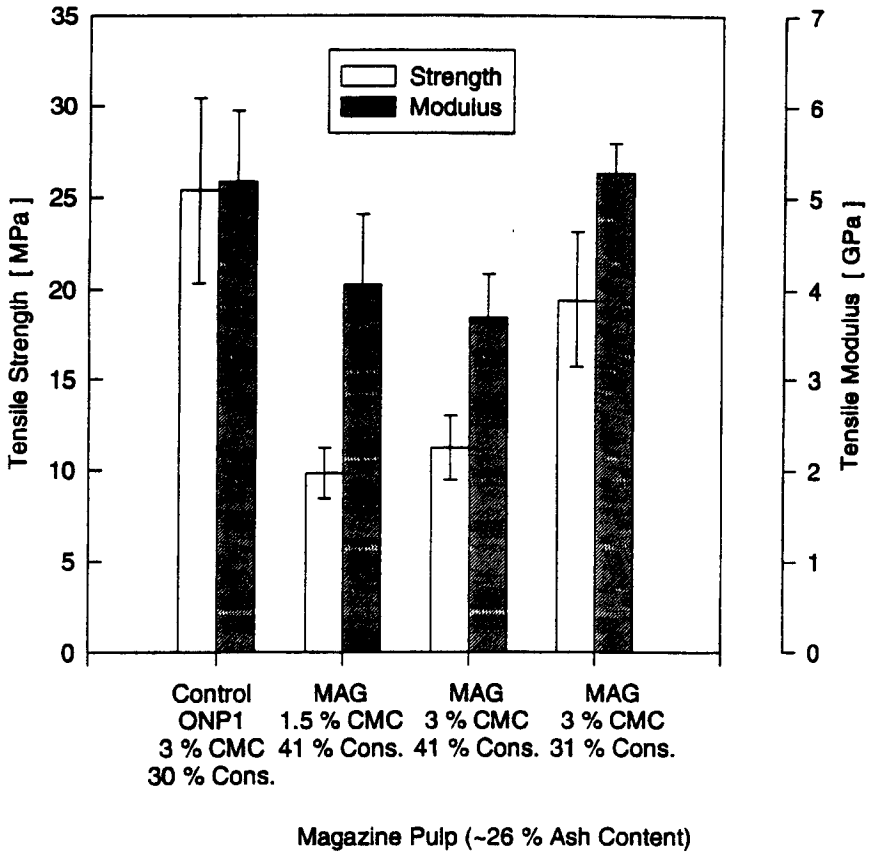

Figure 5.—Tensile strength and tensile modulus for extrudate containing magazine pulp (MAG) at various moisture contents and various Na-CMC additions (runs 4a-c) compared with the control (run 2), unprinted newsprint pulp (ONP1; 3% Na-CMC; 30% consistency).

the extrudate. It is interesting that doubling the addition level of papermaking sludge (about 49% ash content) did not strongly decrease strength and stiffness beyond an initial reduction in strength by 40 percent, and stiffness by 20 percent, at a 10 percent addition level (Fig. 4). For magazine pulp (about 26% ash content) strength and stiffness increased with increasing water and polymer content, despite the better formation obtained at high consistency and low polymer content. This result is consistent with the development of hydrogen bonds in lignocellulosic materials where a high moisture content usually favors the formation of hydrogen bonds. Even with high filler loadings, the mechanical properties of the extrudates compare favorably with commercially available fiberboard and paperboard products (Table 3).

A major impediment to the use of cellulosic fiber composites in structural building applications is the serious loss in tensile modulus and bending stiffness that occurs on their exposure to high humidity and moisture. To overcome this problem, wet-stiffening resins can be used. These resins develop chemical crosslinks that help to preserve the MOE of cellulosics when they are wet and improve dimensional stability by decreasing the moisture sensitivity of the cellulose

Table 3.—Comparison of mechanical properties of pulp extruded composites with those of commercially available wood fiber composites.

Composite	Tensile strength	Elastic modulus
	(MPa)	(GPa)
High density hardboard (12)	27 to 34	5 to 6
Extruded pulp	16 to 26	4 to 6
Laminated paperboard (11)	12 to 15	2 to 3
Medium density fiberboard (11)	11 to 13	2

network to the swelling action of water. Below the fiber saturation point the effect of water in decreasing the MOE of crosslinked cellulosics is quantitatively the same as that of uncrosslinked cellulosics (2). Wet-stiffening arises only from the decrease in the fiber saturation point that the crosslinks create. The role of crosslinks as loadbearing elements is not as important for wet-stiffening as for wet strength. Rather, crosslinks function as swelling restraints to the network, so that a larger fraction of existing hydrogen bonds function to retain a larger fraction of the MOE of the cellulosic material. The same crosslinks that are responsible for dimensional stability and MOE retention are also responsible for the usually observed

reduction in creep. Improvement in mechanical properties in the wet state is often accompanied by a reduction in stretch and tensile energy absorption in the dry state (brittleness). Typical wet-stiffening resins are melamine-formaldehyde (MF) resins, phenol-formaldehyde (PF) resins, and poly(amino)amide-epichlorohydrin (PAE) resins.

At addition levels greater than 5 percent by weight, it was found that PF and MF resins lowered dry tensile strength and stiffness of the extrudates compared to the untreated control (Figs. 6 to 8). This is in line with a homo-crosslinking mechanism, where no additional bonds are created between resin and fiber (3). Formation and dispersion problems were not observed with the PF resins. However, they reduced mechanical properties more than comparable loadings with MF resin. The addition of MF resin caused poorer formation and dispersion at higher addition levels. PAE resin reduced dry strength only slightly and improved dry modulus at any addition level (Fig. 9). This improvement in modulus may be attributed to a synergism developed between PAE and sodium carboxymethyl-cellulose (Na-CMC) (3). It was found that by exceeding a ratio of O.6 Na-CMC/PAE (corresponding to 5% PAE addition in the present experiments), gelation of the Na-CMC occurred and extrusion became impossible due to a loss in lubricating properties of Na-CMC. Formation began to deteriorate already with 2 percent PAE addition. MF, PF, and PAE resins lowered tensile energy absorption in the dry state at all addition levels.

All three resin additives improved wet-strength (Fig. 10) and wet-stiffness (Fig. 11) with increasing addition levels. Figures 10 and 11 also indicate that 20 percent addition is about the limit for useful property improvement. PAE was found to be most effective considering wet-property improvement per unit mass of resin added. However, the ultimate wet-properties were only half of those obtained with the MF and PF resins.

Conclusions

Addition of small amounts of water-soluble polymers to highly concentrated, aqueous pulp suspensions changes their theological behavior such that they can be extruded. These polymers appear to have several important functions in the extrusion process. First, they bind free water, thus avoiding separation of water from pulp fibers under pressure. Second, they add lubricity to the fiber suspension. Third, they aid in floe dispersion. It was found, though not reported here, that very high molecular weight ionic and non-ionic water-soluble polymers such as Na-CMC, hydroxypropylmethyl-cellulose, hydroxyethyl-cellulose, and poly(ethyleneoxide) are effective. Typical addition levels of these polymers are 1 to 3 percent (weight of dry fiber).

After drying, the extruded fiber composites may contain more than 90 percent lignocellulosic fibers, a

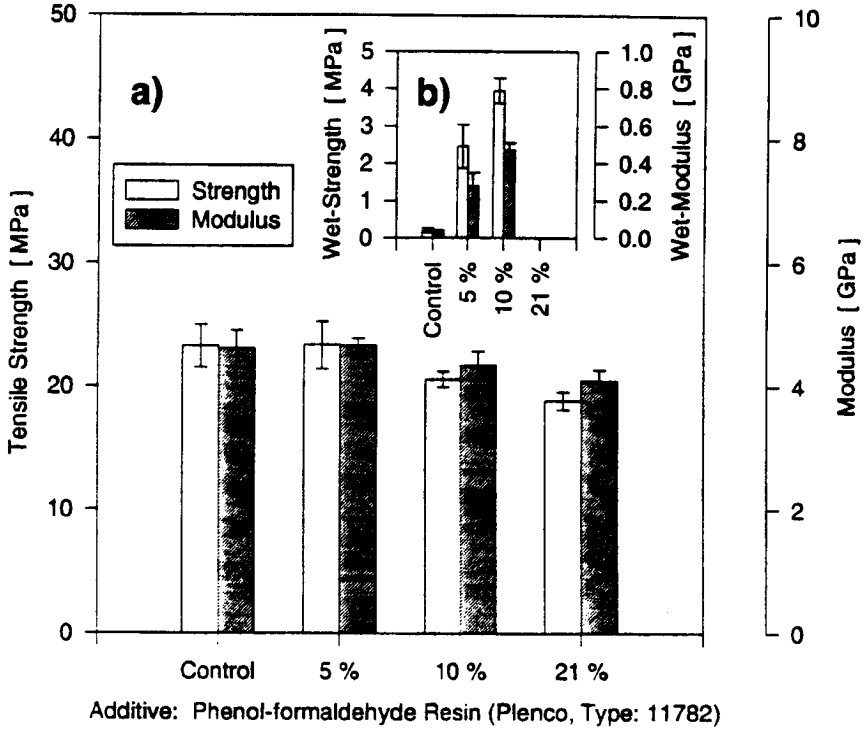

Figure 6.—(a) Dry and (b) wet tensile strength and tensile modulus for pulp extrudate containing 5 (run 6a), 10 (run 6b), and 20 (run 6c) percent phenol-formaldehyde resin (Plenco 11782).

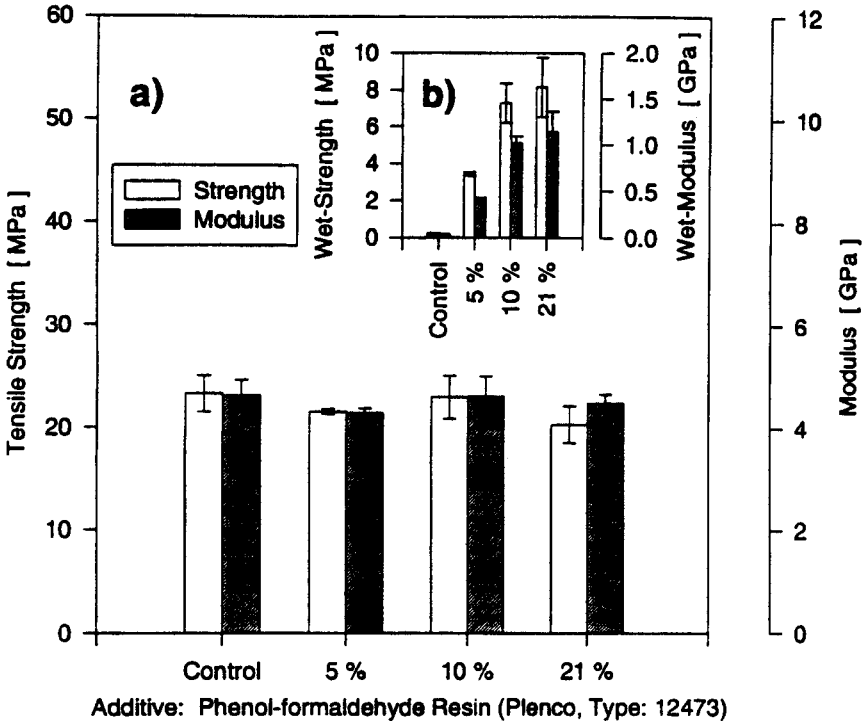

Figure 7.—(a) Dry and (b) wet tensile strength and tensile modulus for pulp extrudate containing 5 (run 7a), 10 (run 7b), and 21 (run 7c) percent phenol-formaldehyde resin (Plenco 12473).

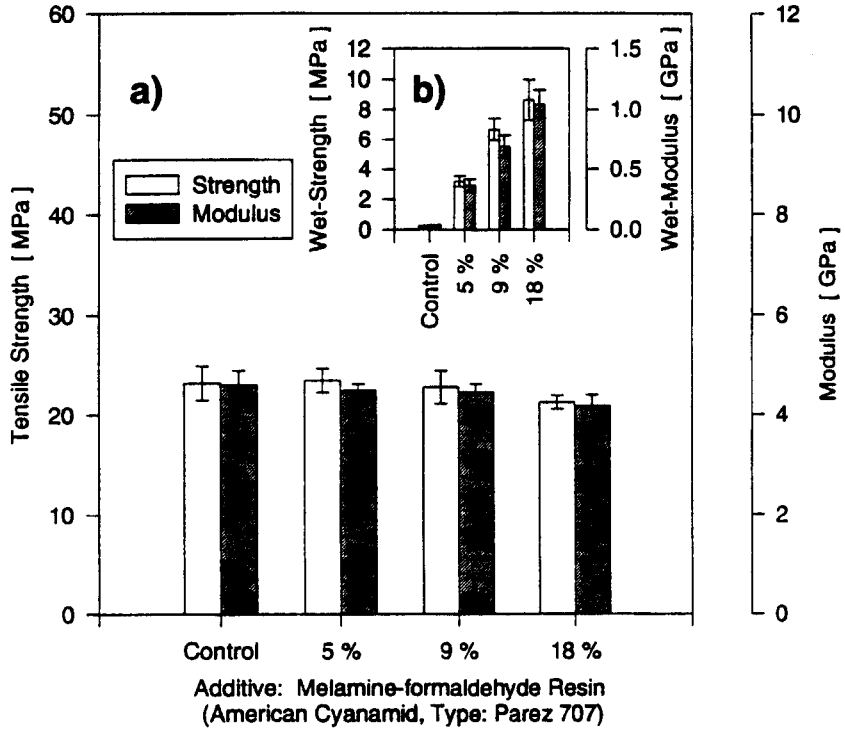

Figure 8.—(a) Dry and (b) wet tensile strength and tensile modulus for pulp extrudate containing 5 (run 8a), 9 (run 8b), and 18 (run 8c) percent melamine-formaldehyde resin (Parez 707).

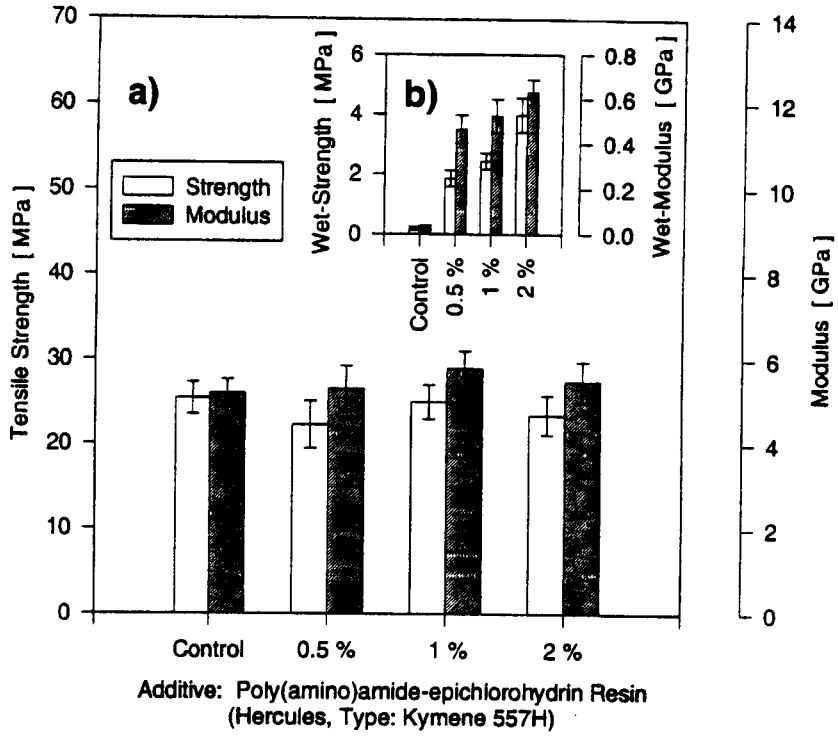

Figure 9.—(a) Dry and (b) wet tensile strength and tensile modulus for pulp extrudate containing 0.5 (run 9a), 1 (run 9b), and 2 (run 9c) percent poly(amino)-amide-epichlorohydrin resin (Kymene 557H).

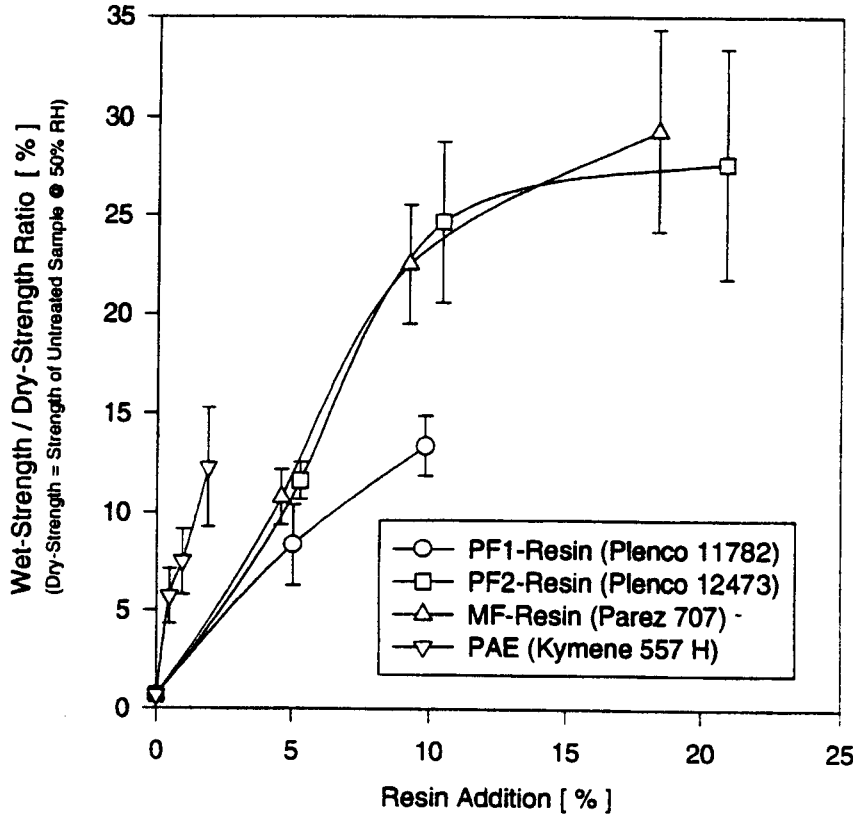

Figure 10.—Wet-strength to dry-strength ratio plotted versus resin addition (PF1= runs 6a-c; PF2=runs 7a-c; MF=runs 8a-c, and PAE=runs 9a-c).

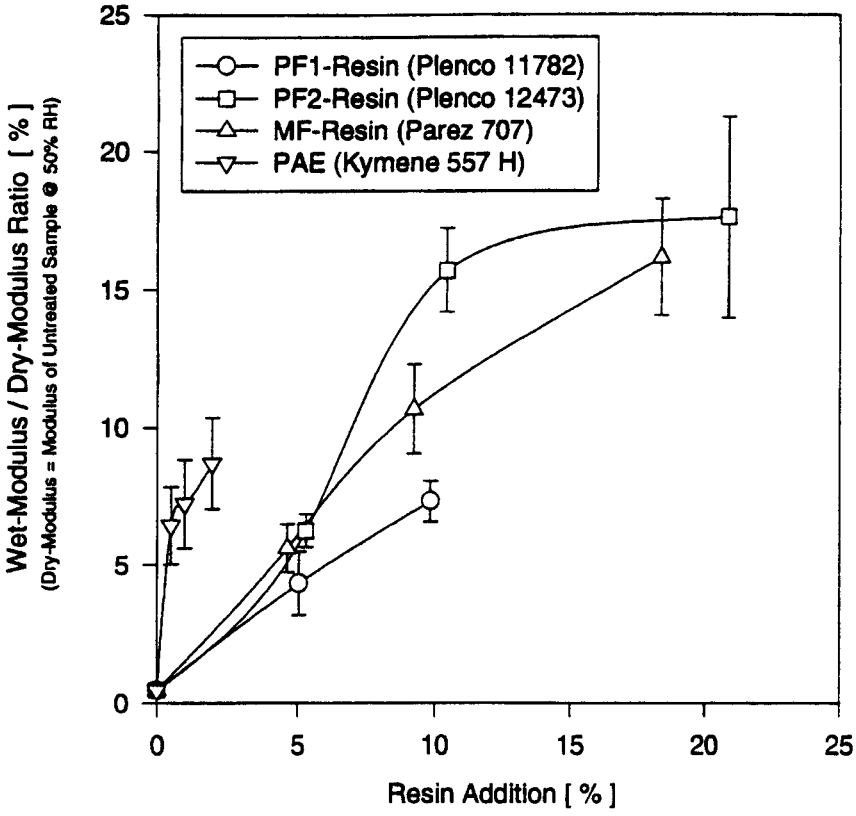

Figure 11.—Wet-modulus to dry-modulus ratio plotted versus resin addition (PF1= runs 6a-c; PF2=runs 7a-c; MF=runs 8a-c, and PAE=runs 9a-c).

much higher fiber fraction than that found in extruded, fiber-filed thermoplastics. The extrusion process is carried out at temperatures below 100°C and low pressures compared with those found in the extrusion of thermoplastics. This is because, in contrast to extrusion of thermoplastics, there is no need to melt the material. Twin-screw extruders were found to be well-suited for processing highly viscous fiber suspensions. Material mixing, fiber dispersion, and forming can all be achieved in a twin-screw extruder where interchangeable screw elements allow customizing the suspension mixing and fiber dispersion. Fiber damage suffered during extrusion processing is small. It was found that fiber length has a strong influence on extrusion behavior; its effect on mechanical properties of the extrudates needs further evaluation.

One important aspect of the present technology is that contaminated fibers from recycled pulp sources and papermill sludges may be used. Pulps containing high filler contents are easily extruded. However, strength and stiffness properties decrease with increasing filler levels.

To obtain structural composite materials, crosslinkable resins, such as PF or MF resins which are commonly available and used in the forest products industry, may be added. These resins ensure that the extrudate does not lose all of its stiffness and strength in high humidity environments. When added in moderate amounts, these resins do not interfere with extrusion processing.

High-consistency pulp extrusion is ideally suited to produce materials with complex cross-sectional shape and tailored material properties. By careful selection of fiber furnish, processing additives, and extrusion conditions, a wide variety of fibers and additives become extrudable. Thus, both structural and nonstructural composite materials can be manufactured. Possible uses may include millwork type applications, high-density oriented products for structural applications, and panel products.

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