

## **Strawboards Bonded with Urea-Formaldehyde Resins <sup>1</sup>**

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### **Abstract**

The conventional strawboard technology employing MDI binders has shown technical and economical disadvantages, specifically, high resin costs and additional costs related to release agents required to avoid sticking of panels to press plates. The adhesion of urea formaldehyde (UF) resin to the straw fiber has been demonstrated in a previous research project using an innovative chemi-thermo-mechanical (CTM) pre-treatment of the straw. This work focuses on the joint efforts to develop a strawboard technology based on UF resins. Three different straw preparation systems, a twin-screw extruder, a pressurized refiner and a combined extruder-refiner treatment, have been chosen to apply intensive shear forces to straw chips in order to overcome the wax and silica layer. With the first two technologies, one-layer panels exclusively bonded with UF have been produced. The board characteristics included excellent appearance, surface smoothness, machinability, edge appearance as well as good bending properties. The internal bond strength values achieved were compatible to particleboard standards. The swelling characteristics did not meet the European standard requirements. With the combined extruder-refiner technology, the UF bonded boards produced showed similarly excellent visual characteristics, bending and internal bond strength values exceeding the MDF standard requirements and swelling figures close to particleboard and MDF standards. By bonding with UF resins, good mat stability, a pressing process without sticking problems and significant resin cost savings can be achieved. The forthcoming development work will be focused on further improvement of the adhesion of straw fibers to produce even higher quality strawboards.

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## Introduction

Recently, there has been a revival of interest to use agricultural fibers due to economic and environmental reasons. Traditionally, in the 50's and 60's, there have been numerous plants based on flax, hemp, and bagasse (Berns and Caesar 1999). Nowadays the interest has been shifted towards palm fibers and cereal straws, primarily from wheat.

The interest in using straw as a raw material is a result of several factors. Straw is an attractive fiber source where wood fiber may not be available or it may allow savings in costs relative to scarce and expensive wood. In other regions with surplus of straw, the usage of straw is promoted for environmental reasons. Plowing straw is not viable in all regions in order to maintain soil balance. Burning straw, which is widely practiced today, creates air pollution and is prohibited in more and more regions. Making panels from straw would contribute favorably to the disposal problem as well as to the overall CO<sub>2</sub> balance as a carbon sink. Farmers can also realize an additional income, being an important factor for promoting the use of straws.

Several publications have discussed the raw material price for straw in comparison to wood fiber and developed optimistic scenarios for the profitability of strawboard plants (Dalen 1999; Bowyer and Stockmann 2001). Typically, the prices for furnish are postulated to be lowest for wood based particleboard and highest for wood based MDF with straw furnish falling between these limits. In the market, the features of strawboard which are most advantageous are, closed panel edges, relatively low density, and the potential for zero-formaldehyde emission levels.

It has been announced that several North American agrifiber board mills have been closed or put into receivership during the recent months (APR 2001). At the present time, only four agrifiber particleboard-type mills are in operation. The above situation clearly indicates that the conventional strawboard technology employing MDI binders is facing problems. Disadvantages such as the high resin costs and the need for appropriate release agents have come critical factors to success.

To date, conventional UF resins have not been utilized in an industrial strawboard production because the straw fiber cells are covered by a layer of wax and silica (Figures 1 and 2). This layer prevents the water based formaldehyde resins, which are widely used for industrial wood panels manufacture, from forming a sufficiently strong bond between the straw fibers. Preliminary studies have shown evidence that the bondability of straw can be improved if its surface structure is opened up and thus made more accessible to wetting by aqueous adhesives such as conventional acid-curing UF resins (Markessini et al. 1997; Mantanis et al. 2000). By applying high shear forces, the wax and silica layer is destroyed and allows bonding with formaldehyde-based resins. For the above-mentioned technical and economical reasons, this work was aimed at developing a strawboard technology that will be based exclusively on UF resin bonding.

## **Straw Preparation Systems Used**

Three different straw preparation systems have been investigated: a twin-screw extruder, a pressurized refiner and a combination of extruder and refiner. These systems apply intensive shear forces to straw chips in order to overcome the aforementioned obstacles. The main difference regarding the shear action is the retention time during preparation. In the twin-screw extruder, the retention time can be a few seconds. In the pressurized refiner system, the straw only remains for fractions of a second between the refiner plates.

### **Twin-screw extruder system**

A twin-screw extruder system was used for dewaxing/defibrating straw under specific chemical, thermal and mechanical conditions. A chemi-thermo-mechanical (CTM) treatment of the straw is thus achieved.

The trials with the twin-screw extruder were carried out in a Clextral BC 45 machine at the facilities of ENSC, Institut Polytechnique de Toulouse, France (Figure 3). The maximum capacity of this machine for straw chips is 50 kg/h (on dry basis).

The twin-screw extruder is a high-shear machine which has the advantage of being a reactor system in which key parameters such as screw profile, retention time, temperature, addition of chemicals can be controlled, and also adjusted to the type of feedstock used (i.e. wheat straw, rice straw, barley straw, rice husk, cotton stalk, etc.). In this work, however, only wheat straw was used. The straw fibers generated show a granulometric distribution that varies depending upon the type of screw profile, liquid/solid ratio and type of chemical reagent used (Figure 4). Additionally, the use of lignin activating agents like NaOH, Na<sub>2</sub>SO<sub>3</sub>, or special wetting agents, which act as lubricants improving the defibration process is possible.

Today, twin-screw extruders are used worldwide in the area of thermoplastics, food processing and to a limited extent in the paper and pulp industry. Within the scope of the project, the maximum throughput figures and the investment costs are under investigation.

### **Pressurized refiner system**

The principal design of the system is well known and comparable to industrial refiners used in modern MDF mills. The trials with the refiner were performed in the Siempelkamp refiner pilot plant (Figure 5) consisting of a feeding bin with presteaming possibility, a plug screw feeder, a digester suitable for handling fibrous raw material, an 22"-Andritz pressurized refiner with flash tube dryer. A blow line blending system may be used to produce under pilot-scale conditions.

### **Combined extruder-refiner system**

This straw preparation system combines the previously mentioned techniques. First, the straw is CTM pre-treated in the twin-screw extruder by using a very soft screw profile at low shear intensity and then it is processed in the pressurized refiner system.

## Trials

### Preparation with the twin-screw extruder

Wheat straw originating from the area of Toulouse was used in these trials. The straw was first hammer milled (a 6 mm sieve was used). The portion of the remaining very fine particles ( $< 0.125$  mm) was limited to less than 4% of the total straw mass. The bulk density of the hammer milled straw was  $50 \text{ kg/m}^3$  (3.12 pcf), while that of fibers generated was around 75 -  $80 \text{ kg/m}^3$  (4.7-5.0 pcf).

The preparation of straw fibers at a constant throughput of 50 kg/h (on dry basis) was carried out with water at a temperature of  $100^\circ\text{C}$  ( $212^\circ\text{F}$ ), while two chemical additives were applied. The first was a wetting agent A (WAA) at a 0.15 % level and the second was a wetting agent B (WAB) at a 0.2 % level (both on dry straw basis). The final moisture content of the fibers at the exit of the machine was 90-95 % (on dry basis). The specific energy at these conditions was approx. 230-250 kWh/dry ton.

The straw fibers were then dried in a circulating-air oven. A moisture level of 3 % was adjusted. No screening/sieving was carried out after drying.

The pH of the straw fibers was neutral. Their buffering capacity was comparably high at 8.8 - 9.2 (Table 1). For comparison, spruce MDF fibers typically have a buffering capacity of 4.4 - 4.5 ml of 0.1 N HCl.

From the two types of extruder prepared straw, five one-layer straw panels measuring  $0.46 \times 0.46$  m, 16.9 mm thick ( $1\frac{1}{2}$  by  $1\frac{1}{2}$  ft.,  $\frac{2}{3}$  in.) were manufactured at the facilities of the Adhesives Research Institute (ARI Ltd.), Thessaloniki, Greece, in order to investigate the relation between type of CTM treatment and strawboard performance. The parameters selected for board production are given in Table 2. The panels were manufactured at two target densities –  $730 \text{ kg/m}^3$  (45.6 pcf) and  $760 \text{ kg/m}^3$  (47.4 pcf) - the press temperature was  $190^\circ\text{C}$  ( $374^\circ\text{F}$ ). The strawboards were evaluated for density, mechanical properties (IBS, MOR), 24 hour swell and formaldehyde release potential using the perforator method (EN 120). The results are given in Table 3.

The bending strength values obtained were between  $19.8 - 21.5 \text{ N/mm}^2$  (~3,000 psi), while the internal bond strength was  $0.50 - 0.56 \text{ N/mm}^2$  (~77 psi); both of these property values are compatible to the common particleboard requirements, although they are close to those of the European MDF standards. There were no significant differences between the two chemically different treatments. The swelling figures of both panel types are exceeding the limit of 15 % and therefore do not meet the corresponding European requirements.

Nonetheless, it should be stressed here that the above-mentioned results have come out at the completion of the optimization phase of the extruder preparation system. Presently, the emphasis is being shifted towards the fortification of the plain UF resin system, and particularly on the incorporation of special cross-linking agents or melamine, as well as other resin additives.

### **Preparation with the pressurized refiner system**

To study the refining characteristics of straw, the following trials were performed:

- three different pre-steaming and digesting conditions (trial A)
- two different pre-treatments with alkaline chemicals (trial B)
- refining speed (trial C)

The trial variations are given in Table 4. The straw fibers produced were characterized chemically and by screen analysis. Screening with an Alpine air jet screen proved to be suitable to characterize the fibers (Table 5). Fiber pH values and buffer capacities were high in relation to other raw materials as has also been found in the extruder trials. The addition of alkaline chemicals increased the buffer capacity even more.

Visual examination of the fibers showed clearly that straw tends to render a high amount of non-fibrous dust and a considerably high content of shives. Shives create poorer panel surfaces and create adhesion problems with water based resins.

Within trial A, fibers of different granulometry were produced by adjusting the refiner gap. Panels were pressed and tested. The basic production parameters are given in Table 6. Very fine fibers characterized by a high dust content have not been suitable for producing UF bonded panels. The panels tended to blister even at low densities. The maximum densities achieved have only been around  $620 \text{ kg/m}^3$  (38.7 pcf).

Medium coarse fibers rendered higher panel densities up to  $\sim 700 \text{ kg/m}^3$  (43.7 pcf). Standardized to a panel density of  $675 \text{ kg/m}^3$  (42.1 pcf), the MOR values were approximately  $20 \text{ N/mm}^2$  (2,900 psi) and the MOE approx.  $2,800 \text{ N/mm}^2$  (406,000 psi). The European MDF standard MOR EN 622-5 could just be reached, while the bending MOE was comfortably exceeded. The IBS varied around  $0.45 \text{ N/mm}^2$  (65.3 psi) and 24 hour swelling figures were 16 – 17 %. In both cases, the standard requirements for MDF were not reached. Testing coarser fibers did not show any improvement; therefore, for the test trials B and C only medium coarse fibers were produced.

The impregnation with sodium hydroxide changed the fiber quality only to a minor extent. The screen analysis has shown nearly identical granulometry for non-treated and 0.2 %-treated straw fibers. After addition of 0.5 % NaOH, the fiber granulometry was slightly coarser. The dust portion decreased from 14 % to 8 %, while the 50 %-passing was shifted from 0.55 mm to 0.75 mm screen mesh.

The characteristics of panels from both sodium hydroxide treatments were nearly identical. The MOR values standardized to  $675 \text{ kg/m}^3$  density (42.1 pcf) have been approximately  $18 \text{ N/mm}^2$  (2,600 psi) and the MOE approx.  $2,300 \text{ N/mm}^2$  (335,000 psi). The internal bond strength was improved to approx.  $0.55 \text{ N/mm}^2$  (80 psi) and the swelling figures were between 15 and 20 % with no significant difference between both treatments.

The differences compared to the panels produced from untreated straw may be attributed to the slight change in the density profile. A small increase of mat moisture content before pressing from approximately 12 % to 13.5 % might have shifted the raw density profile

toward higher core density. Therefore, the bending properties have decreased somewhat and internal bond strength increased. A significant influence of the chemical pre-treatment could not be found.

The important influence of the density profile is demonstrated by the panels made with different pressing programs in trial C. The density profiles are shown in Figure 6. In comparison to other raw materials, straw fibers are very sensitive to adjustments of the pressure program. From a given straw fiber batch pressed under identical conditions of temperature and maximum pressure, panels with large differences in density profiles and mechanical properties were obtained. For panel 1 with a high surface density and a low core density/average density ratio of 69 %, the IBS was only 0.35 N/mm<sup>2</sup> (51 psi) and MOR was as high as approximately 30 N/mm<sup>2</sup> (4,350 psi). In panel 2 with an optimized density profile and a core density/average density ratio of 86 %, the MOR was slightly reduced to 25 N/mm<sup>2</sup> (3,625 psi), while the IBS was raised to 0.50 N/mm<sup>2</sup> (72.5 psi).

The intention of decreasing the refiner speed in trial C was to reduce milling effects on the straw fibers. In fact, the amount of shives was reduced indicating that the shear treatment was increased. Better panel properties were therefore expected.

Unfortunately, the results were superimposed by a difference in straw characteristics due to change in the straw origin. Summarizing, the standardized (675 kg/m<sup>3</sup> density) panel properties were improved with an IBS of 0.5 N/mm<sup>2</sup> and a bending strength of 25 N/mm<sup>2</sup>, compared to alkaline-treated and non-treated straw. The maximum achievable densities have been increased by approximately 50 kg/m<sup>3</sup> (3.12 pcf). All swelling figures exceeded 20 % with no improvement compared to sodium hydroxide treated straw.

### **Preparation with the combined extruder-refiner system**

Pre-chipped French wheat straw bigger than 2 mm screen mesh was extruded using a very soft profile at low shear intensities (around 70-80 kWh/dry ton). Two different chemical pre-treatments have been tested. First, sodium hydroxide with subsequent neutralization by boric acid and second, pure water was added. The pre-treated straw was shipped to the Siempelkamp pilot plant and processed directly for refining.

The refining was performed after pre-steaming at 80 °C and digesting for 1 ½ min. The refiner pressure was set to 6 bar. The refined fibers were dried, blender-resinated and used to form one-layer mats. The boards were produced according to the conditions given in Table 7.

Panels have been produced at densities ranging from 675 to 725 kg/m<sup>3</sup> without press factor optimization. The panel properties are given in Table 8.

The MOR figures fulfill the requirements of the European MDF standard EN 622-5. The ANSI MDF standard A208.2-1994 requires a bending strength of 25 N/mm<sup>2</sup> (3,625 psi) which will be fulfilled with densities exceeding 730 kg/m<sup>3</sup> (45.6 pcf). The MOE requirements of both standards are easily met with values exceeding 3,000 N/mm<sup>2</sup> (435,000 psi).

The IBS values ranged from 0.65 N/mm<sup>2</sup> (94 psi) for a density of 675 kg/m<sup>3</sup> to 0.82 N/mm<sup>2</sup> (109 psi) at 725 kg/m<sup>3</sup> density. There were no significant differences between both preparation methods. The standard requirements of 0.55 N/mm<sup>2</sup> (EN) and 0.60 N/mm<sup>2</sup> (ANSI), respectively, were met. The swelling figures of the two panel types differed significantly. Fairly high swelling figures of 27 % for water-treated material and 35 % for chemical treated material were found for panels of a lower density. With increasing density, a significant decrease in the swelling was obtained. At a density of 725 kg/m<sup>3</sup> (45 pcf), 24 hour swelling figures of 15 % and 24 % were respectively achieved.

### **Summary and Conclusions**

Although the relatively high pH and buffer capacity of straw slows down the curing speed of acid-curing resin, panels from 100% wheat straw bonded exclusively with UF resin have been produced successfully.

Application of high shear forces in a refiner and an extruder made the bonding of straw fibers exclusively with UF resins possible. Different straw preparation systems have been tested. Extruding straw with a twin-screw extruder or refining with a pressurized refiner have rendered panels with characteristics comparable to standard particleboard, except for their swelling properties. Producing panels from refined fibers needs several precise adjustments of pre-treatment and refining conditions as well as a good density profile program. By combining these measures, significant improvements with regard to the maximum achievable density as well as to the panel properties have been made.

A combined treatment by extruder and refiner has rendered improved panel properties comparable to MDF. Although, the swelling figures were improved, the 24 hour swell standards for MDF and particleboard could not yet be fulfilled. Special care has to be taken in relation to the inherent timely and regional variability of straw.

By bonding with UF resins, good mat stability, a pressing process without sticking problems and significant resin cost savings can be realized. Further development work should focus on improvements of the bondability of straw fibers to produce even higher quality strawboards, specifically lower swelling figures. Industrial pilot trials will be performed.

### **Acknowledgements**

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**Table 1.**—pH and buffer capacity\* of extruder treated straw.

Material	pH	Buffer capacity
WAA- treated straw	7.0	8.8
WAB- treated straw	7.0	9.2
Spruce MDF fibers	4.1	4.5

\* ml 0.1N HCl to reach a pH of 3.0

**Table 2.**—Parameters selected for the production of strawboards from extruder treated straw fibers.

Parameter	Level
Board thickness	16 mm (0.63")
Resin addition	12 % E2 UF
Press temperature	190°C (374 °F)
Paraffin (on dry straw)	1 %
Hardener (on dry resin)	2 %
Press factor	15 sec/mm

**Table 3.**—Properties of one-layer strawboards produced from WAA- and WAB- extruder treated straw fibers.

Types	Density kg/m <sup>3</sup> (pcf)	IBS N/mm <sup>2</sup> (psi)	MOR N/mm <sup>2</sup> (psi)	24h swell %	HCHO mg/100g (EN 120)
WAA-1	760 (47.4)	0.53 (76.9)	21.5 (3,118)	17.3	16.2
WAA-2	730 (45.6)	0.50 (72.5)	19.9 (2,886)	17.9	17.0
WAB-1	762 (47.6)	0.56 (81.2)	20.3 (2,944)	16.2	14.2
WAB-2	735 (45.9)	0.52 (75.4)	19.8 (2,872)	16.7	14.9

**Table 4.**—Refining parameters.

Trial	Pre-steaming temperature (°C)	Cooking time (min)	Refiner speed (min <sup>-1</sup> )	Chemical addition (%)	Straw origin*, year
A 1	without	3	standard	without	D, 1999
A 2	without	1 ½	standard	without	D, 1999
A 3	80 °C, 10 min	1 ½	standard	without	D, 1999
B 1	80 °C, 10 min	1 ½	standard	0.2 % NaOH	D, 1999
B 2	80 °C, 10 min	1 ½	standard	0.5 % NaOH	D, 1999
C	80 °C, 10 min	1 ½	reduced	without	F, 2000

\* D: Germany, F: France; 80 °C = 176 °F

**Table 5.**—Screen analysis by Alpine air jet screening of straw fibers of medium coarseness from different pre-treatments.

Trial	screen passing, cumulative %				
	0.2 mm (0.008")	0.5 mm (0.0196")	1.0 mm (0.0393")	2.0 mm (0.080")	3.15 mm (0.124")
A 3 fine	33	67	95	100	100
A 3 medium	14	42	72	94	100
A 3 coarse	8	28	58	87	98
B 1 medium	14	47	76	92	99
B 2 medium	8	31	65	84	96
C medium	8	43	82	98	100

**Table 6.**—Production parameters for strawboard from refiner material.

Parameter	Level
Board thickness	16 mm (0.63")
Resin addition	13 % E2 UF
Press temperature	195 °C (383 °F)
Maximum specific pressure	250 N/cm <sup>2</sup> (362 psi)
Press factor	15/18 sec/mm

**Table 7.**—Production parameters for strawboards from extruded-refined material.

Parameter	Level
Board thickness	16 mm (0.63")
Resin addition	13 % E2 UF
Press temperature	195 °C (383 °F)
Maximum specific pressure	250 N/cm <sup>2</sup> (362 psi)
Press factor	18 sec/mm

**Table 8.**— Properties of strawboards produced from extruded-refined straw fibers.

Agents	Density	MOR	MOE	IBS	24h swell
	kg/m <sup>3</sup>	N/mm <sup>2</sup>	N/mm <sup>2</sup>	N/mm <sup>2</sup>	%
	(pcf)	(psi)	(psi)	(psi)	
NaOH / H <sub>3</sub> BO <sub>3</sub>	675 (42)	20.8 (3,016)	3,210 (465,500)	0.65 (94)	35.5
NaOH / H <sub>3</sub> BO <sub>3</sub>	725 (45)	25.7 (3,727)	3,640 (528,000)	0.82 (119)	24.2
H <sub>2</sub> O	675 (42)	21.3 (3,089)	3,110 (451,000)	0.66 (96)	27.0
H <sub>2</sub> O	725 (45)	23.8 (3,452)	3,580 (519,000)	0.74 (107)	15.0

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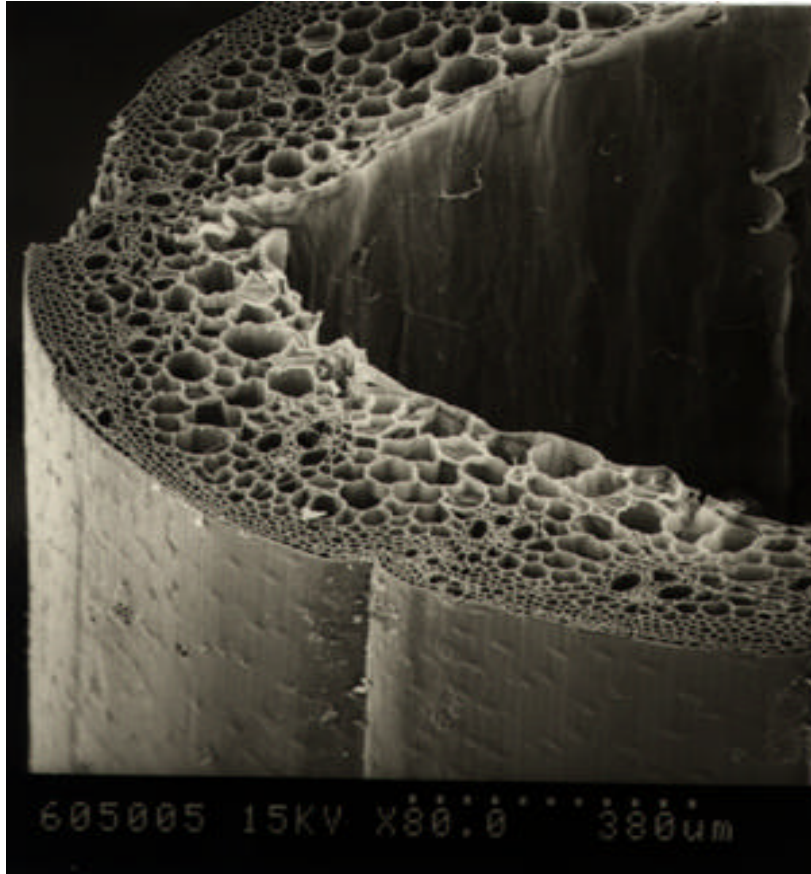
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**Figure 3.**—Pilot scale twin-screw extruder (Clextral BC 45).

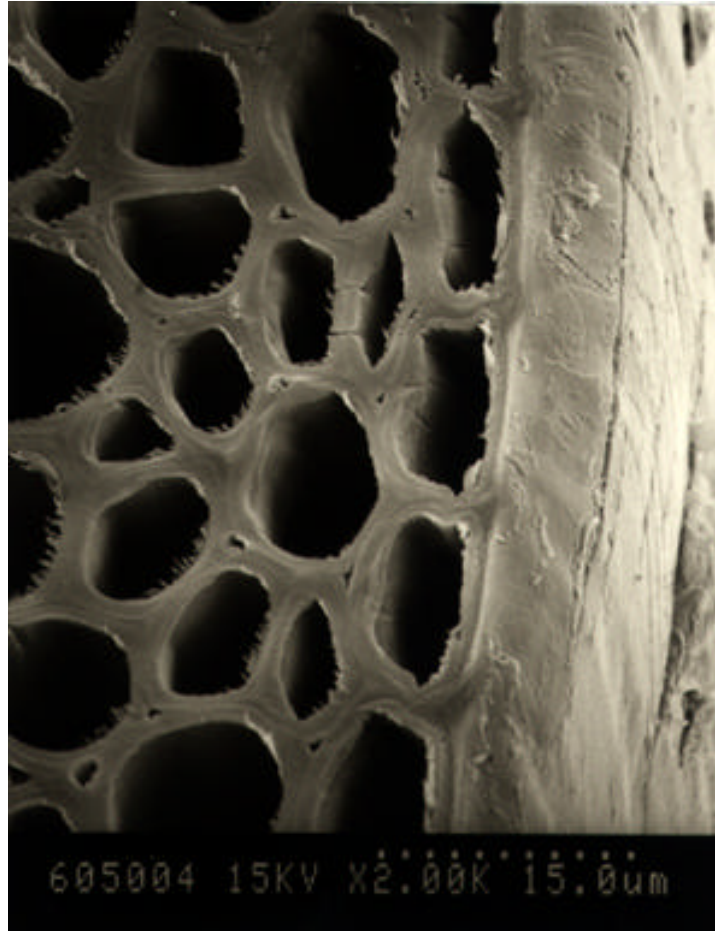
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**Figure 5.**—The Siempelkamp refiner pilot plant.

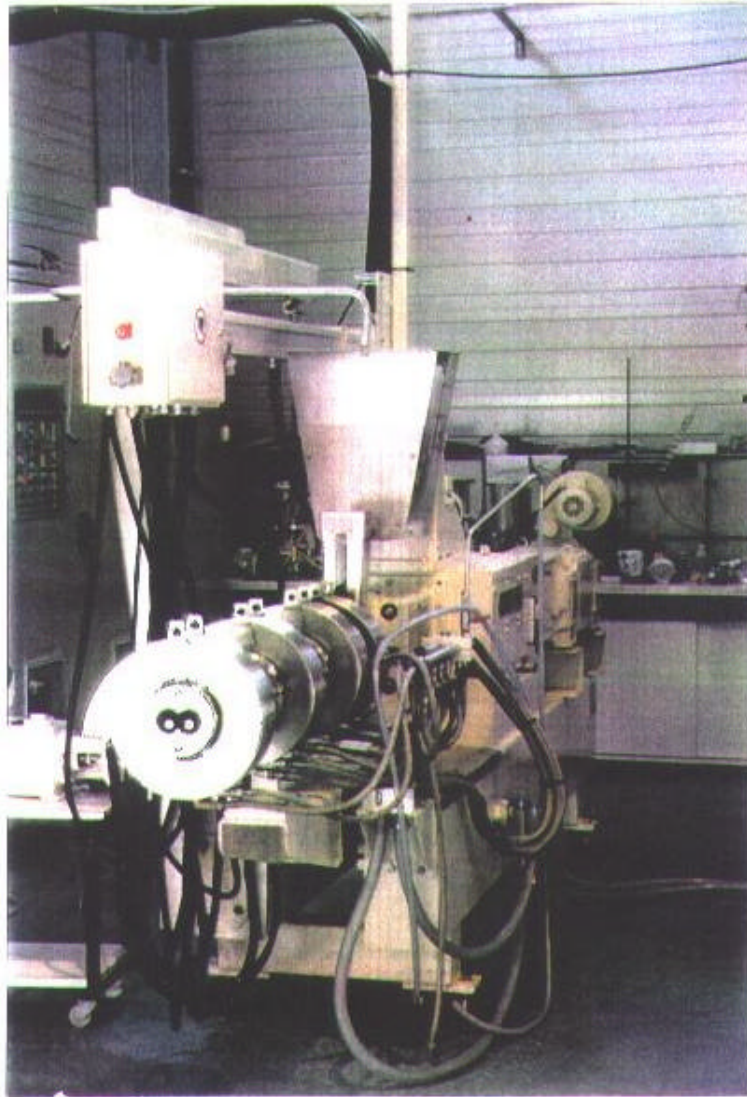
**Figure 6.**—Density profiles of strawboards.



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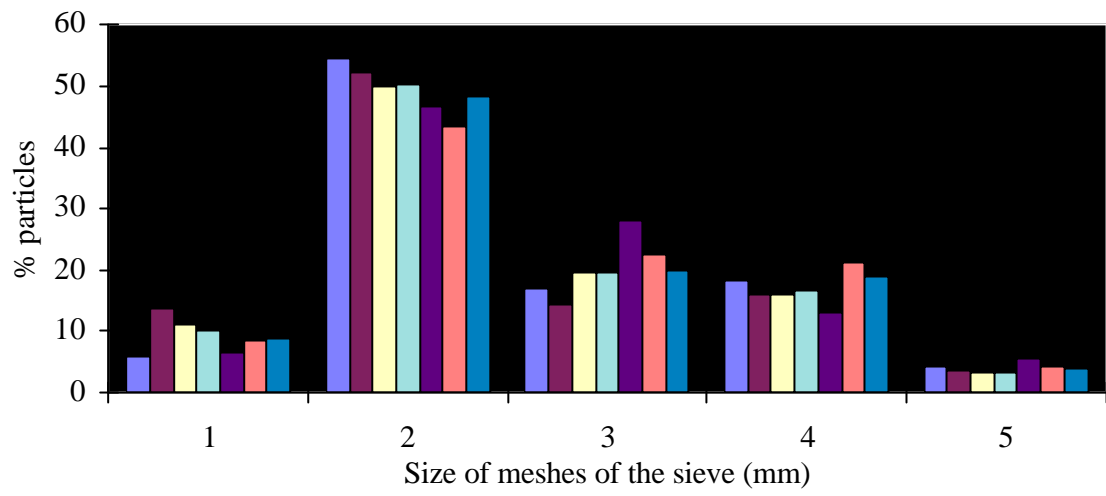


**Figure 2.**— Electron micrograph of a straw waxy/silica layer.



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**Figure 4.**—Granulometric distribution of extruded straw fibers.

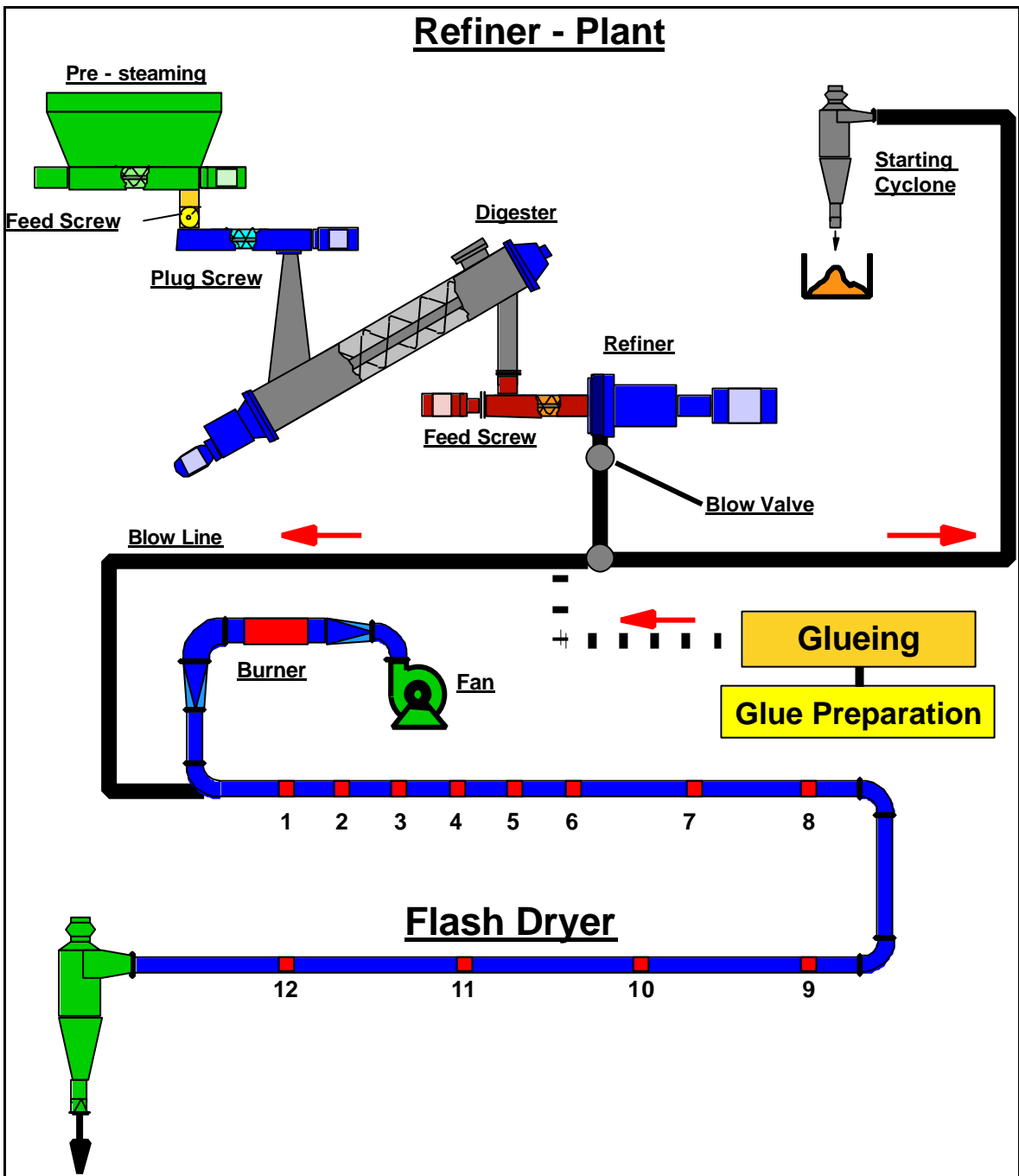
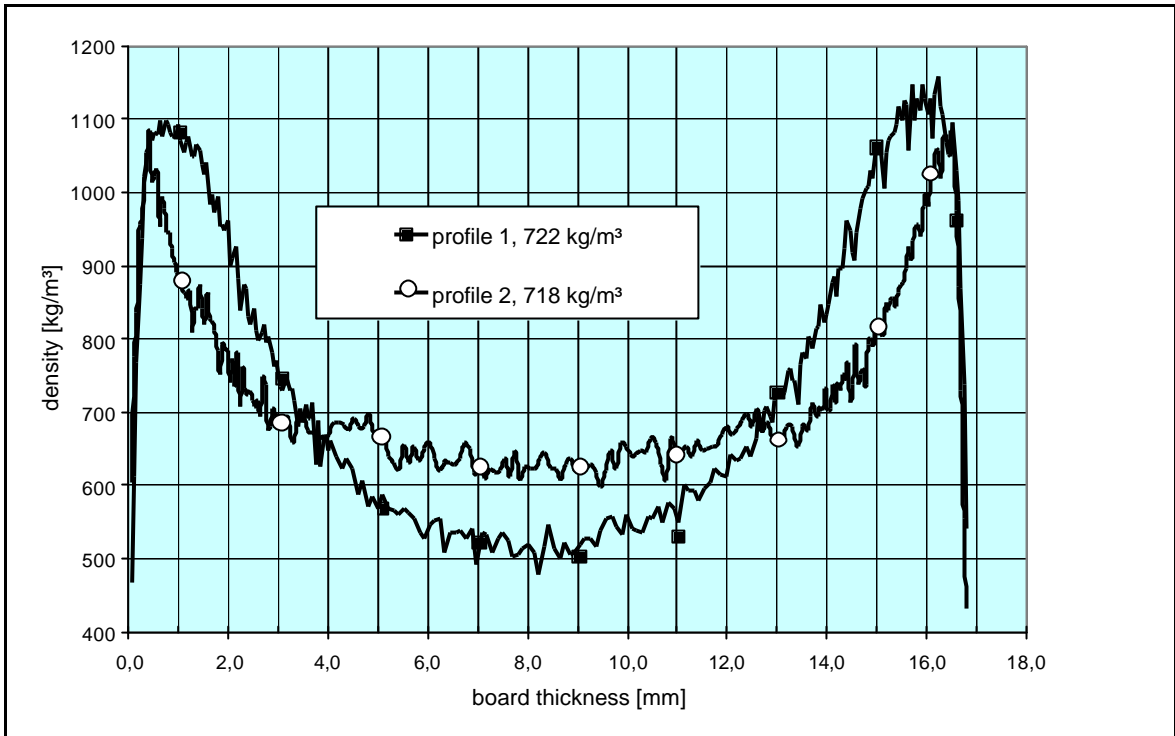


Figure 5.—The Siempelkamp refiner pilot plant.



**Figure 6.**—Density profiles of strawboards.