STATUS REPORTS

to the

PAPER PHYSICS

PROJECT ADVISORY COMMITTEE

Volume I

March 23, 1999
TO: MEMBERS OF THE PAPER PHYSICS PROJECT ADVISORY COMMITTEE

Attached for your review are the Status Reports for the projects to be discussed at the Paper Physics Project Advisory Committee meeting. The Program Review is scheduled for Tuesday, March 23, 1998, at 8:00 a.m. - 5:00 p.m. and the PAC Committee Meeting will be held on Wednesday, March 24, 1999 from 8:00 a.m. to 12:00 p.m.

Please note that the meeting is being held at the Institute of Paper Science and Technology.

We look forward to seeing you at this time.

Sincerely,

John Waterhouse
Acting Director
Fiber and Paper Physics Division

JFW/djh

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* The dates in ( ) indicate the final year of the appointment.
## PROGRAM REVIEW AGENDA

### Seminar Room

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<td>Opening Remarks and Antitrust Statement</td>
<td>John Waterhouse &amp; Doeung Choi</td>
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<td>8:20 a.m. - 8:40 a.m.</td>
<td>Welcome from the Vice President of Research</td>
<td>Gary Baum</td>
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<td>8:40 a.m. - 9:30 a.m.</td>
<td>Project F020 Fundamentals of Dimensional Stability</td>
<td>Doug Coffin</td>
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<td>9:30 a.m. - 10:20 a.m.</td>
<td>Project F026 Fundamentals of Accelerated Creep</td>
<td>Chuck Habeger</td>
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<td>10:20 a.m. - 10:35 a.m.</td>
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<td>10:35 a.m. - 11:10 a.m.</td>
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<td>Wayne Robbins</td>
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<td>11:10 a.m. - 12:00 p.m.</td>
<td>Project F023 Fundamentals of Micromechanics of Fiber Networks</td>
<td>Martin Ostoja – Starzewski</td>
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<td>12:30 p.m. - 1:20 p.m.</td>
<td>Project F024 Improving the Refining of Chemical Pulps</td>
<td>John Waterhouse</td>
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<td>1:20 p.m. - 2:10 p.m.</td>
<td>Project F025 Fundamentals of Interfiber Bonding</td>
<td>Hiroki Nanko</td>
</tr>
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<td>2:30 p.m. - 3:20 p.m.</td>
<td>Project F007 On-line Measurement of Paper Properties</td>
<td>Maclin Hall</td>
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<td>3:20 p.m. - 4:10 p.m.</td>
<td>Project F008 Fundamentals of Acoustic Radiation Pressure</td>
<td>Pierre Brodeur</td>
</tr>
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<td>4:10 p.m. - 5:00 p.m.</td>
<td>Project F031 Paper Stiffness Versus Papermaking Variables</td>
<td>Pierre Brodeur</td>
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### Dimensional Stability and Creep

### Micromechanics, Refining and Bonding

### Ultrasonics
PAPER PHYSICS
PROJECT ADVISORY COMMITTEE MEETING

MARCH 24, 1998

Institute of Paper Science and Technology
Atlanta, Georgia

COMMITTEE DISCUSSIONS AGENDA

Room 173

8:00 a.m. - 8:20 a.m.
- Antitrust Statement/Confidentiality Statements
- Next Meeting Date
- Review of Agenda

8:20 a.m. - 8:35 a.m.
- RAC Liaison’s Overview of Research Lines/Roadmaps
- Research Lines/Roadmaps
- Research Ideas List
- Project Review Critique Forms

8:35 a.m. - 10:00 a.m.
- Subcommittee Discussions
  Project F020 Dimensional Stability
  Project F026 Accelerated Creep
  Project F023 Micromechanics
  Project F024 Fiber Properties
  Project F025 Bonding
  Project F007 On-line Measurement of Paper Properties
  Project F008 Acoustic Radiation Pressure
  Project F031 Paper Stiffness VS Papermaking Variables

10:00 a.m. - 12:00 p.m.
- Full Committee Discussion
- Close Meeting
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ON-LINE MEASUREMENT OF PAPER PROPERTIES

STATUS REPORT

FOR

PROJECT F007

Mac Hall
Ted Jackson
Andy Brown

Institute of Paper Science and Technology
500 10th Street, N. W.
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DUES-FUNDED PROJECT SUMMARY

Project Title: ON-LINE MEASUREMENT OF PAPER PROPERTIES
Project Code: ONLIN
Project Number: F007
PAC: Paper Physics
Division: Fiber and Paper Physics
Project Staff: Mac Hall, Ted Jackson, Andy Brown
FY 98-99 Budget: $94,845
Allocated as Cost Share in Cooperative Agreement No. DE-FC02-95CDE41156
Time Allocation: 20%
Supporting Research:
Related External Funding has been provided by DOE/Office of Industrial Technology

RESEARCH LINE/ROADMAP

10. Energy Performance
12. Sensors and Process Control

PROJECT OBJECTIVE
This project focuses on the fundamentals of paper stiffness measurement using ultrasonics and the relationships of ZD and in-plane stiffnesses to on-machine process parameters. It supplements the Cooperative Agreement project to develop commercially viable sensors and instrumentation capable of providing ultrasonic measurements in the in-plane and thickness directions of the paper web as it is being made on the paper machine.

PROJECT BACKGROUND
The variations of the elastic stiffnesses of paper with refining, fiber orientation, wet pressing pressure, wet straining (draws), and drying restraints have been studied and reported (Baum et al., 1984; Habeger and Baum, 1986). These studies have demonstrated that elastic stiffnesses are sensitive to changes in furnish and to changes in various process parameters. Measurement on the paper machine of the elastic stiffnesses in both the in-plane and thickness directions along with the basis weight, moisture, and caliper of the web should provide means to continuously monitor product quality and data to control the paper manufacturing process.

Confidential Information - Not for Public Disclosure
(For IPST Member Company's Internal Use Only)
The instrumentation should be applicable to most grades of paper, but will be particularly beneficial to the heavier weight grades, packaging paper and paperboard, because "strength" properties are of primary importance for these products.

The instrumentation has the potential of providing:

1) Improved First Quality
   A. Quicker Grade Changes
   B. Less Culls Due to CD "Strength" Variations
   C. Less Downgraded Tonnage During the Run

2) Fiber Savings
   A. Reduce Fiber Usage While Maintaining "Strength" Targets
   B. Use Less Expensive Fiber Sources While Maintaining "Strength" Targets
   C. Reduce Ring Crush Variability Creating Opportunity to Operate at Lower Ring Crush Targets

3) Energy Savings
   A. Minimize Refiner Loads While Maintaining "Strength" Targets
   B. Reduce Total Steam Requirements Through Better Drainage and Less Fiber to Dry

4) Increased Production
   A. Increase Machine Speed while Maintaining "Strength" Targets

A 1000-tpd machine producing 350,000-tpy uses approximately 4.725 trillion Btu/year. For each 1% of production that is substandard and reprocessed at 13.5 million Btu/ton, the 1000-tpd machine wastes 47.25 billion Btu/year. Assuming this technology may limit substandard production to 2%, the energy savings would be 94.5 billion Btu/year or $378,000 (@ $4/million Btu) annually for the 1000-tpd machine.

Further energy savings should result from optimum utilization of refining and drying (Lantz and Chase, 1988). Refining requires approximately 200 kWh/ton or 2.1 million Btu/ton (1 kWh = 10,500 Btu). For a 1000-tpd machine, the annual energy usage for refining is approximately 70.0 million kWh or 735 billion Btu. Assuming the optimization
of refining could reduce the energy required by 10%, this would be equivalent to savings of 73.5 billion Btu or $294,000 annually.

By decreasing refining, drainage is improved, requiring less steam for drying the web. The steam required for drying is equivalent to approximately 8 million Btu/ton, or 2.8 trillion Btu/year for a 1000-tpd machine. If the moisture of the paper entering the dryer were reduced by 1.0%, the dryer steam required would be reduced by about 3%. This would save an additional 84 billion Btu/year or $336,000 annually.

Summary for a 1000-tpd paper machine:

Annual Energy Use = 4.725 trillion Btu = $18,900,000 @ $4/million Btu

* Reprocessing savings:
  * Electricity 38.5 billion Btu
  * Steam 56.0 billion Btu
  Total reprocessing savings 94.5 billion Btu = $378,000

* Refining savings:
  * Electricity 73.5 billion Btu = $294,000

* Drying savings:
  * Steam 84.0 billion Btu = $336,000

Total potential savings for 1000-tpd machine:

* Electricity 112.0 billion Btu = $448,000
* Steam 140.0 billion Btu = $560,000
  @ $4/million Btu = $1,008,000

Total potential savings/year 252 billion Btu/year = 5.3%

While designing and developing instrumentation to demonstrate on-machine ZD measurements in a mill, this project has supported various related studies in the laboratory. These studies provide background and support for verifying on-machine sensor performance and on-machine measurement and process relationships. Some examples are:
Demonstrated repeatable measurements of short-range variations of MD, CD, and ZD ultrasonic velocities for cross-machine samples of machine-made papers.

Ultrasonic velocity measurements of CD strips show large short-range variations that appear similar to the nonuniformities, streaks, and "dry-line fingers" observed on the forming table.

Examined various correlation techniques to determine relationships between ultrasonic velocity measurements and compressive strength measurements, Ring Crush (RC), and short-span compressive strength (STFI).

Developed system to make high-resolution measurements of basis weight (using the ABB Beta gage) and of ultrasound velocity in the ZD (using fluid-filled wheels) for paper samples mounted on the laboratory web handler.

Demonstrated sensitivity of ZD ultrasonic measurements to wet pressing, refining, and calendering using samples made on Herty pilot machine.

SUMMARY OF RESULTS (March 98 - February 99)

1. In-plane sensor operational on machine in mill.

2. ZD system tested at ABB and first installed on machine in mill in July 1998.

3. Mechanical interface of wheels to web while scanning successfully demonstrated with control of wheel speed and steering for a run exceeding 500 miles.

4. Electronics system and software to record and display ZD ultrasonic transit time and pulse energies developed and operated in mill.

5. Collected ZD data for total of 30 hours on September 8-11, scanning on approximately 480 miles of web. Displayed data as "waterfall" display with up to 100 scans in display for transit time, reflected energy at transmitter, reflected energy at receiver, or absorbed energy.

6. FactNet 4.5 software (Pacific Simulation) was used to analyze relationships of ultrasonic measurements, process variables, and product properties.

PROJECT GOALS FOR FY 99-00

Verify on-machine ultrasonic sensor measurements by comparison with laboratory instrument measurements.
Combine ZD data from "stand alone" E-box with the ABB data processing and display system. This will provide ZD data for operator and for the mill's PI data recording system.

Determine relationships between on-machine ultrasonic velocity measurements and papermaking process variables, e.g., refining, wet pressing, wet straining, and calendering. Emphasize the sensitivity of ZD ultrasonic measurements to process variables.

PROJECT DELIVERABLES:

1. Verification of on-machine sensor measurements.
2. Relationships between on-machine ultrasonic measurements, papermaking process variables, and paper properties.

PROJECT SCHEDULE:

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<td>Combine ZD data with ABB data processing and display system</td>
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<td>2.</td>
<td>Verify on-machine sensors: (1) In-plane; (2) ZD</td>
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<td>Determine correlation of ultrasonic measurements with mill grade specifications</td>
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OBSERVATIONS AND DISCUSSION

In-Plane Ultrasonic Measurements

Confidential Information - Not for Public Disclosure
(For IPST Member Company’s Internal Use Only)
The on-machine in-plane sensor measures the CD longitudinal velocity \((V_{CD})\) and the shear velocity \((V_{SH})\) in the cross direction. We are also interested in knowing the MD longitudinal velocity \((V_{MD})\). This can be calculated in the following way.

We start with the relationship (Baum et al., 1981)

\[
G = \rho (V_{SH})^2 = \frac{[(E_{MD})(E_{CD})]^{1/2}}{2[1 + (\nu_{MD}\nu_{CD})^{1/2}]}
\]  

For anisotropy ratios less than 3, the in-plane Poisson ratio, \((\nu_{MD}\nu_{CD})^{1/2}\), was found by Baum et al. to be essentially constant with a mean value of 0.293. Substituting this into Eq. 1 gives

\[
\rho (V_{SH})^2 = 0.387 [(E_{MD})(E_{CD})]^{1/2}
\]  

where \(E_{MD} = \rho (V_{MD})^2 (1 - \nu_{MD}\nu_{CD})\) and \(E_{CD} = \rho (V_{CD})^2 (1 - \nu_{MD}\nu_{CD})\)

Although small, the Poisson ratio should not be ignored in the relationship between \(E\) and \(V\). Substituting these into Eq. 2,

\[
(V_{SH})^2 = 0.387 (V_{MD})(V_{CD}) (1 - \nu_{MD}\nu_{CD})\), \text{ where } \nu_{MD}\nu_{CD} = (0.293)^2
\]  

or

\[
(V_{SH})^2 = 0.354 (V_{MD})(V_{CD})
\]  

Thus, the MD longitudinal velocity \((V_{MD})\) may be calculated if one has a measure of the CD longitudinal velocity \((V_{CD})\) and the shear velocity \((V_{SH})\),

\[
V_{MD} = 2.82 \frac{(V_{SH})^2}{V_{CD}}.
\]  

The IPST robot for in-plane measurements has been used to measure the \(V_{MD}\), \(V_{CD}\), and \(V_{SH}\) along the CD of CD strips of several linerboard grades. The relationships of the
measured velocity values were found to be consistent with a proportionality constant of approximately 2.8.

**Status of On-Machine Demonstration**

The Institute has a Cooperative Agreement with the U.S. Department of Energy’s Office of Industrial Technologies (DOE-OIT) for a project to develop and demonstrate commercially viable on-line ultrasonic sensors. The project includes systems with the ability to make both in-plane and out-of-plane (ZD) ultrasonic measurements.

IPST is prime contractor with cost-share participation by ABB Industrial Systems Inc., Columbus, Ohio; the Herty Foundation, Savannah, Georgia; and the Georgia-Pacific Mill in Cedar Springs, Georgia. We are in the fifth year of a 5-year development and testing program.

An AccuRay® 1190™ System with a Smart Platform™ 1200 has been installed on the web handling system in the laboratory at IPST and on a pilot machine at the Herty Foundation. A similar system is installed at the Georgia-Pacific Cedar Springs Mill. It has been operating on Georgia-Pacific’s paper machine #1 (PM#1) since April 1997. The sensor carriage in each of these scanners contains state-of-the-art basis weight, moisture, temperature, and caliper sensors. The sensor carriage in each of these scanners also accommodates the installation of in-plane and ZD ultrasonic sensors. ABB has built and installed in-plane sensors at IPST, at Herty, and at the Cedar Springs Mill.

IPST has designed and built a ZD ultrasonic sensor using transducers mounted in fluid-filled wheels. After an initial test at Herty, it was decided to redesign the wheel mounting modules so that the wheels could be steered during scanning. The redesigned modules were tested on a web handler in ABB’s Lab in Columbus, Ohio, on July 6-9, 1998. The system was first install on PM#1 at the Cedar Springs Mill during outage on July 28, 1998. Following short tests on machine, the modules have been removed to correct problems and reinstalled. Access to the sensor carriage on the
scanner has been limited primarily to the one-shift-a-month outage of PM#1 for maintenance.

A number of changes have been made to the ZD system since it was first installed in the mill: an air conditioner was added to the E-Box; cooling air was directed at the wheels; the wheels were overhauled with new bearings, seals, and connectors; silicone rubber tires were substituted for polyurethane tires; the fluid in the tires was changed to a special fluid for high-temperature ultrasonic coupling; two co-axial cables and two air hoses were installed in both the top and the bottom auxiliary flex tracks of the scanner for communication with the ZD modules; the solenoid air valves were replaced; and the modules were rewired. The position of the in-plane and the ZD sensors in the carriage was switched so that the center of gravity is closer to the center of support in the scanner frame. The ZD modules are now downstream and the in-plane system upstream in the sensor carriage.

Detailed plans have been prepared for both step tests and end-of-reel tests to determine relationships between in-plane stiffness measurements, sheet properties and process variables for PM#1. The process variables to be manipulated, the scanned profiles to be collected, and the tests to be performed on collected samples are outlined below.

**Process Variables to be Manipulated:**

1. Basis Weight
2. Machine Speed
3. Base Refiner Load
4. Rush/Drag
5. Broke Flow
6. DLK Flow

**Scanned Profiles to be Collected:**

1. Basis Weight
2. Percent Moisture
3. Caliper
4. Temperature
5. Shear Velocity Squared
6. CD Longitudinal Velocity Squared

Tests to be Performed on Collected CD Strip Samples at G-P:

1. CD Ring Crush
2. L&W TSI CD, TSI MD, & MD/CD Ratio
3. Mullen, felt side & wire side
4. STFI
5. Basis Weight
6. Percent Moisture
7. Caliper
8. MD Tensile
9. MD and CD Tear

In addition to the CD strips to be collected for testing at the mill, two CD strips for IPST will be obtained for each test reel. A complimentary set of off-line measurements will be performed at IPST.

The data will be transferred to Excel spreadsheets and analyzed using FactNet 4.5 software. The time stamps for the ABB, G-P, and IPST systems need to be the same. Care will be required to match samples with the profiles and data to ensure spatial alignment between collected data and actual strips. The trials are scheduled for late February and the first set will be with 55 lb/1000 ft² (USP 120) linerboard. The second set at a later date will be 69 lb/1000 ft² linerboard.

The above plan has been designed primarily for evaluating the in-plane system. However, IPST will collect scanned data with the "stand alone" ZD system during the tests. Combining the ZD data with the in-plane and process data recorded in the PI system will be cumbersome and potentially inaccurate in matching time and position. However, it should provide useful information.

In order to provide a better basis for evaluating the sensitivity of ZD measurements to process changes and to determine the potential value of ZD data to the operator, a second ZD system design is currently in progress. This system will provide ZD data to the ABB Smart Platform to be processed along with the ABB data. ZD data will then be available to the operator and to the PI system.
The plan for combining the ZD system with the ABB scanner, 1190 and Advant display involves no additional electronics on board the ZD modules in the sensor carriage. A separate electronics cabinet (E-Box) with air conditioning would continue to be used. This E-box will operate the ZD modules and collect ZD ultrasonic data similar to the present system.

The ZD E-box will forward two streams of preprocessed ZD data to the end column of the Smart Platform. The data will then be handled by the ABB system similar to the way data from the other sensors in the scanner are processed and forwarded for storage and display.

REFERENCES


FUNDAMENTALS OF ACOUSTIC RADIATION PRESSURE

STATUS REPORT

FOR

PROJECT F008

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Mee Choi
Jimmy Jong
Brian Pufahl

Institute of Paper Science and Technology
500 10th Street, N. W.
Atlanta, Georgia 30318
DUES-FUNDED PROJECT SUMMARY

Project Title: FUNDAMENTALS OF ACOUSTIC RADIATION PRESSURE

Project Code: FARPE
Project Number: F008
PAC: Paper Physics
Division: Fiber and Paper Physics Division

Project Staff
Faculty/Senior Staff: Pierre Brodeur
Staff:
  Joseph Gerhardstein (Associate Engineer)
  Feler Bose (Assistant Engineer)
  Mee Choi (Postdoc)
  Jimmy Jong (Postdoc)
  Brian Pufahl (Assistant Engineer)

FY 98-99 Budget: $107,746
Allocated as Matching Funds: $107,746

Time Allocation
Faculty/Senior Staff: Pierre Brodeur (12%)
Support:
Joseph Gerhardstein (19%), Feler Bose (24%),
Mee Choi (5%), Brian Pufahl (10%), Jimmy Jong (15%)

Supporting Research
M.S. Students: None
Ph.D. Students: None

Related External Funding:
- Project #4183 "Acoustic Separation Technology" - Sponsored by Department of Energy and Beloit Corporation
- Project #4190 "Closed Water Treatment in Pulp Mills Using A Dual Flocculation/Ultrasonic Clarification Method" - Sponsored by State of Georgia Traditional Industry Program in Pulp and Paper - In collaboration with Prof. Yulin Deng (IPST Engineering and Papermaking Division)

RESEARCH LINES/ROADMAPS: 4, 7, 11, 12, and 13

PROJECT OBJECTIVES:
- Investigate fundamentals of acoustic radiation pressure (ARP) effects on fiber suspensions
- Demonstrate selected pulp and paper industry-related applications of acoustic radiation pressure
- Determine economic viability of acoustically-based processes

IPST Confidential Information - Not for Public Disclosure
(For IPST Member Company’s Internal Use Only)
PROJECT BACKGROUND:

Project F008 was initiated in July 1992 to explore various effects of acoustic radiation pressure on water-suspended wood pulp fibers and other particulate matter. Since then, considerable work has been accomplished to develop a structured research program, develop specialized test equipment (e.g., 450 l/min acoustic separation system), perform preliminary demonstrations (e.g., transverse deflection of fiber suspensions subjected to traveling and standing ultrasonic wave fields in laminar and turbulent flows, acousto-optical method to determine wet compactibility of fiber suspensions), and seek external funding to supplement IPST’s funding effort. Combined funding from all sources enables the following nonproprietary activities:

- Acoustic separation of vessel elements and hardwood fibers (#F008)
- Development of automated image analysis method to quantify the presence of vessel elements in hardwood furnishes (#F008)
- Demonstration of dual chemical flocculation/ultrasonic clarification method (#4190)
- Theoretical/numerical analysis of acoustic radiation force acting on wood pulp fibers under different flow conditions (#4183)
- Separation technology development (#4183)
- Safety analysis (#4183)
- Economical, technical, and energy assessments of technology (#4183)
- Technology transfer (#4183)

As indicated above, project F008 currently focuses on the proof-of-concept of vessel/hardwood fiber separation.

SUMMARY OF RESULTS (March 98 - March 99):

- Much of the emphasis has been on the improvement of a semi-automated image analysis test method to measure the size distribution of vessels in hardwood furnishes. However, as superior as it is believed to be when compared to conventional testing procedures used in the industry, the test method failed to provide repeatable results when no power was applied to the transducers. Sample preparation remained problematic and time consuming. This is very unfortunate because it prevented us from making any significant progress in the vessel separation method itself. Different series of experiments were performed as a function of consistency, frequency, acoustic intensity, flow rate, and divider blade position. Each time, several weeks were needed...
to process samples collected in just a few hours, only to conclude that not only was there some evidence of separation, but also inconsistent trends. It was decided in January 1999 to put on hold the separation application and refocus the project starting in FY99-00. For the remainder of FY98-99 (February to June), it is planned to determine shortfalls of the test method for vessel analysis and validate it using different furnishes and measurements obtained using a conventional approach (vessel counting using a microscope). The goal is to transfer the test method to IPST Member Companies by June 1999.

- A teaming agreement between IPST and Beloit Corporation was finalized in January 1999. This is a significant milestone, which will drive the future of project F008, which is the cost share to DOE Agenda 2020 “Acoustic Separation Technology” project (IPST project 4183).

PROJECT GOALS FOR FY 1999-2000:

- Project goals for FY99-00 will be redefined in collaboration with Beloit Corporation. A new set of goals will be proposed to the Paper Physics PAC during the review meeting in March. These goals will focus on some fundamental issues of acoustic radiation pressure vs. fiber suspensions.

PROJECT DELIVERABLES:

- Semi-automated image analysis test method for vessel counting and vessel size distribution (June 1999).

PROJECT SCHEDULE:

<table>
<thead>
<tr>
<th>Tasks</th>
<th>FY 97-98 (Jul-Aug)</th>
<th>FY 98-99 (Jul-Aug)</th>
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<tr>
<td>Equipment Purchasing</td>
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<tr>
<td>System Upgrading</td>
<td>---</td>
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<tr>
<td>Microscopy/Staining Work</td>
<td>---</td>
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<tr>
<td>Vessel Analysis Method</td>
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<tr>
<td>Prelim. Trials (Training)</td>
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<tr>
<td>Vessel Separation Experim.*</td>
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<tr>
<td>Application Assessment*</td>
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<td>---</td>
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<tr>
<td>Final Report</td>
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* Vessel separation experiments and application assessment put on hold in January 1999.
1. Introduction

This status report presents an overview of the progress accomplished so far in project F008. First, in order to appreciate current research activities, background information from the early days of the project is provided in Section 2. Section 3 provides a description of the experimental setup used to demonstrate the separation of vessel elements and hardwood fibers. Section 4 delves into the economic analysis of the acoustic separation technology. Section 5 provides an introduction to the theoretical/modeling research effort on fibers interacting with an ultrasonic wave field. Section 6 provides a brief review of accomplishments about the separation of vessels and hardwood fibers. Section 6 also shows the results of the runs done in the last few months as well as the summary of research activities concerning the development of the automated image analysis method for vessel detection.

2. Background

Project F008 began in July 1992. This project was set up to study the fundamentals of acoustic radiation pressure effects on fiber suspensions and investigate potential applications in the pulp and paper industry. In essence, when water-suspended wood pulp fibers interact with an ultrasonic wave field, they are subjected to an acoustic force which results in fiber migration [Brodeur, 1991; Brodeur, 1989; Garceau et al., 1989; Dion et al., 1988]. There is also an acoustic torque which reorients fibers. Both acoustic force and torque are nonlinear effects. These effects are very attractive because they enable the noncontact mechanical manipulation of flowing fibers. Although several studies have shown that fluid-suspended particles can agglomerate...
when submitted to traveling or stationary ultrasonic wave fields, very little is known in the context of prolate spheroids (cigar-shaped particles) or cylindrical particles. Also, the study of fiber suspensions interacting with an ultrasonic wave field in laminar and turbulent flows is not documented in the literature. An understanding of acoustic manipulation of fibers at large flow rates is of primary importance.

Very early in the project, a basic experimental apparatus was built to investigate the acoustic force on nonmoving and flowing fiber suspensions. A special acoustic cell mounted in an unpressurized vertical pipe of square cross section was devised to deflect an input stream of fibers interacting with a traveling ultrasonic wave field (unidirectional field) normal to the flow direction. The cell is composed of a 2 x 10 cm active area piezoelectric ceramic transducer operating at 150 kHz and a sound absorber of equivalent area. In this arrangement, fibers flow against gravity and deflect toward the absorber position when the sound field is turned on. The transducer-absorber separation distance is 2 cm.

Preliminary experimental observations confirmed that the acoustic force could potentially be used to separate/fractionate fibers based on fiber width [Brodeur, 1994]. Consequently, an in-flow divider blade was installed above the acoustic cell in such a way as to separate highly deflected fibers (coarse fibers) from weakly deflected or undeflected fibers (slender fibers and/or fines). This would lead to at least two output streams: a coarse fiber-enriched stream and a coarse fiber-depleted stream. Literature/patent searches did not reveal evidence of past work in this area. A patent application was filed by IPST in 1996 and was granted in Sep. 1998.
In a particular series of experiments using the basic experimental setup, rayon fibers of constant width/variable length and constant length/variable width were tested as a function of acoustic intensity and flow rate at 0.05% consistency [Ma, 1995]. Rayon fibers were used as they best simulate wood pulp fibers. Separation efficiency was indirectly determined by measuring the cleanliness efficiency of the weakly deflected fiber output stream, i.e., by analyzing the percentage of fibers remaining in the “clean stream.” Cleanliness efficiency [CE] is defined as follows:

$$CE = 100(1 - f_C/f_F)$$  \[2-1\]

where $f_C$ and $f_F$ refer to the percentage by weight of solids oven dried for the clean and feed streams, respectively.

Results supporting the hypothesis that fiber migration is a function of fiber width are shown in Figures 2-1 and 2-2. While the cleanliness efficiency increases as a function of intensity for all test samples (at constant flow rate), indicating an increased migration rate from the transducer to the absorber, the cleanliness efficiency increases more as a function of fiber width in Figure 2-1 and is less affected by fiber length in Figure 2-2.
Figure 2-1. Cleanliness efficiency as a function of acoustic intensity for constant width/variable length rayon fibers. Fiber dimensions were determined a priori using a microscopy method.

Figure 2-2. Cleanliness efficiency as a function of acoustic intensity for constant length/variable width rayon fibers.

In a similar set of experiments, a Bauer-McNett classifier was used to separate two fractions of never-dried hardwood fibers (sample A, average fiber length: 0.78 mm; sample B, average fiber length: 1.10 mm). The cleanliness efficiency was determined for samples A and B and a mixture labeled M (25% A - 75% B; predicted fiber length: 1.02 mm). Results are presented in Figure 2-3. Clearly, the efficiency increases as a function of fiber length (related to fiber width in this case).
Figure 2-3. Cleanliness efficiency for samples of never-dried hardwood fibers.

![Diagram showing cleanliness efficiency with fiber length and acoustic intensity][1]

Figure 2-4. Pulp thickening effect [2 cm (horizontal) x 10 cm (vertical)].

Another series of observations was obtained using softwood fibers under zero-flow conditions to study the compactibility of fiber suspensions. Figure 2-4 shows a compacted fiber suspension for an initial pulp consistency of 1% (*white indicates water and black indicates fiber*). The acoustic intensity is 0.3 W/cm². One can easily see a pulp thickening effect. A new test method, acoustic wet fiber compactibility, has been proposed to control the refining process and predict the apparent density of paper [Brodeur and Runge, 1996].
Using a more efficient transducer (> 90% power efficiency conversion from electrical to mechanical power), video recordings were obtained for different acoustic intensity levels at 70 L/min (18 gal/min) using never-dried softwood fibers (at max. pump capacity; Re ≈ 6000 - turbulent regime). Results are shown in Figure 2-5 (black indicates fiber). Pulp consistency is 0.1%. One can see that fiber deflection (toward the left) significantly increases as a function of intensity (shown as a thickening of the fiber mat (black) on the left side of the cell). At 4 W/cm², the intensity is already too large and deflected fibers bounce back.

**Figure 2-5.** Video recordings under turbulent flow conditions [2 cm (hor.) x 10 cm (vert.)].

Other test results can be summarized as follows:

(1) Migration rate for shives is significantly larger than that for fibers;
(2) Migration rate for earlywood fibers is larger than that for latewood fibers;
(3) Observations using OCC fibers show that the fiber fraction migrates but the fines do not; and
(4) Observations using mixtures of fibers and ink particles attached to air bubbles show that the air bubbles are less affected by the acoustic force, thus providing a means to enhance the removal of fibers in flotation deinking.

Experiments were also conducted using a standing ultrasonic wave field. This field is obtained by a reflector instead of an absorber. The reflector was made of stainless steel. Providing that the separation distance between the transducer and the reflector corresponds to \( n (\lambda/2) \), where \( \lambda \) is the acoustic wavelength (10 mm at \( f = 150 \) kHz in water), interference between the transmitted and reflected wavefronts produces a standing wave field pattern. Since the transducer-reflector separation distance was 2 cm, four nodal pressure planes were created. Observations using wood pulp fibers showed that the fibers quickly migrate toward pressure planes when the acoustic field is turned on, thus leading to the formation of four parallel agglomeration planes. This is illustrated in Figure 2-6.

![Agglomeration Planes](image)

**Figure 2-6.** Agglomeration of 5.9 mm rayon fibers at 1% consistency in a standing wave field. Four layers are produced. The Reynolds number is approximately 500.
Technical limitations (small size of equipment, consistency and flow limitations) in the quest to perform realistic separation experiments using existing equipment led to the decision in 1996 to design and fabricate a larger-scale experimental setup. The separation system was functional in March 1997, but with limited capabilities. It was used to gather preliminary measurements on the separation of vessel elements and hardwood fibers [Oakland, 1997]. Additional equipment to drive transducers and determine power consumption was purchased during the fall of 1997. Details of this apparatus are reported in the next section.

In parallel to the development of the larger-scale experimental setup and the vessel-fiber separation study, an investigation of ultrasonic refining of pulp suspensions using a 20-kHz ultrasonic processor was performed by John Blanz [Blanz, 1997]. The most interesting finding was that high power ultrasonics can potentially be used to decurl fibers.

Finally, a patent was issued on the separation technology in September 1998 [Brodeur, 1998].
3. Ultrasonic Separation Apparatus (Joseph Gerhardstein)

The separation apparatus currently used for experiments is shown schematically in Figures 3-1 and 3-2.

![Flow system for the laboratory prototype acoustic separator (drawing not to scale).]

Briefly, the flow cell consists of a 1.9-meter flow development section, an acoustic section containing three 5 x 10-cm piezoelectric transducers above it, and is topped by an atmospheric pressure mechanical separation system. The flow cell is rectangular in cross section, with one dimension fixed at 5 cm, and the other variable from 5-15 cm (in 5-cm steps). A 450 l/min (maximum) variable speed centrifugal pump is used to produce flows in the cell with Reynolds numbers between 400 and 140,000. A 500-liter stock tank is used to feed the pump, and a pair of 200-liter stock tanks are on the output of the flow cell (one on each side). The cell is typically run in a close loop mode, where the stock flows from the 500-liter tank, through the pump, into the flow cell where
it is acoustically processed, then out into one of the 200-liter tanks and back to the 500-liter tank (it can also be run in batch mode where the sample is entirely collected in the 200 liter tanks). Samples are usually taken from the 200-liter tanks while the cell is running in closed loop mode. A mechanical flow straightener was added to the 1.9-meter flow development section to solve some problems with non-uniform flow when using the maximum wall spacing of 15 cm.

![Diagram](image)

**Figure 3-2.** Schematic diagram of the laboratory prototype AST system (*drawing not to scale*). Separation using a traveling wave field (unidirectional) is shown.

Figure 3-3 shows the electrical setup for the acoustic cell. Three separate channels are used, one for each transducer, to provide flexibility in how the system is run. A computer controls three function generators via a GPIB network. These function generators can be phase locked so that the waves are synchronized. The output from the function generators is fed into three power amplifiers, which boost the signal strength to the amplitude necessary to drive the transducers. Various amplifiers with powers up to 1500 watts and bandwidths from 20 kHz to 10 MHz are used. As the
output from the power amplifiers is at 50 ohms, and the transducers have a much larger input impedance, a set of matching networks is used to perform the transition. Inline power meters are positioned between the matching networks and transducers to provide closed-loop control of the power going to the transducers.

![Diagram of electrical setup for the acoustic cell.](image)

**Figure 3-3.** Electrical setup for the acoustic cell.

A picture of the transducers is shown in Figure 3-4. Transducers of three different frequencies are currently used in the flow cell: 1.5 MHz, 150 kHz and 60 kHz. All are 5 x 10 cm in shape and are easily interchangeable. The transducers are designed to handle temperatures up to 100°C.
Figure 3-4. Transducers used in flow cell, 1.5 MHz (left), 150 kHz (center) and 60 kHz (right).

Heating in the transducers and matching networks is caused by inefficient power conversion. In order to determine where electrical power is being lost, an oscilloscope with high frequency current and voltage probes is used to measure power along the electrical path from the wall outlet to the transducer as shown in Figure 3-5. Measurements will be done between the wall outlet and the power amplifier (amount of power the amplifier consumes), between the power amplifier and the matching network (amount of power output by the amplifier), and between the matching network and the transducer (power delivered to the transducer).
Figure 3-5. Measurement of electrical power usage from wall to transducer using new oscilloscope and high frequency probes. Oscilloscope is shown measuring power between Power Amplifier and Matching Network.

Electrical-to-acoustic efficiency of the transducers is measured using an energy balance method, as shown in Figure 3-6. An oversized absorber is hung underwater from a balance. The transducer to be evaluated is rigidly supported above the absorber. When power is applied to the transducer, the radiation pressure is transmitted to the absorber and read on the balance. A calibration factor of 6.8 g per watt of acoustic power is used. An optional anti-streaming membrane can be inserted between the transducer and absorber to eliminate streaming effects.
Figure 3-6. Energy balance method for determining transducer efficiency. (a) Conceptual schematic showing transducer, absorber, balance and water tank. (b) Picture of actual setup.

4. Economy Analysis of Acoustic Separation Technology

This section looks at a specific application for the Acoustic Separation Technology, dewatering of stock. Experiments were performed on IPST’s laboratory scale separation system using a bleached softwood kraft pulp. The pulp was run through the acoustic apparatus at various flow rates (from 0.05 to 0.4m/s) and various consistencies (0.01 to 1.0% C), and the angle of deflection of the pulp under the acoustic field was measured as a function of acoustic intensity. From these data, the amount of acoustic power ($P$) to deflect the pulp flowing at 1000 gallons per minute (GPM) halfway across a pipe was calculated using the following formula:

$$P = IA_1$$  \hspace{1cm} (4-1)
where $\ell$ is the acoustic intensity (power per unit area going into the transducer) and $A_t$ is the area of the transducer, which can be written as the product of the length ($\ell$) and width ($w$) of the transducer:

$$A_t = \ell w$$

(4-2)

For an arbitrary acoustic section depth $d$, and a pulp deflection angle of $\theta$, the length of the acoustic section to achieve a 50% deflection, i.e., to double stock consistency, is given by

$$\ell = \frac{d/2}{\tan(\theta)}$$

(4-3)

The width of the acoustic section can be calculated from

$$Q = v A = v d w$$

(4-4)

or

$$w = \frac{Q}{v d}$$

(4-5)

where $Q$ is the volume flow rate, $v$ is the flow speed in the acoustic section and $A$ is the cross section of the acoustic section ($d w$). Hence, substituting equations 4-3 and 4-5 into equation 4-2, the area of the transducer ($A_t$) can be expressed as

$$A_t = \left( \frac{d/2}{\tan(\theta)} \right) \left( \frac{Q}{v d} \right) = \frac{Q}{2 v \tan(\theta)}$$

(4-6)

and an expression for the power necessary to deflect the pulp half way across the acoustic cell is then given by combining equations 4-1 and 4-6:

$$P = \frac{I Q}{2 V \tan(\theta)}$$

(4-7)

Note that this equation is independent of the depth $d$ of the acoustic section.
Figure 4-1 shows surface mesh plots of the data at initial consistencies of 0.1% and 1.0%. The power necessary to double the consistency for a nominal flow rate of 1000 GPM (equation 4-5) is shown as a function of the flow speed and the acoustic intensity.

**Figure 4-1.** Power consumption to double stock consistency as a function of flow speed and acoustic intensity for 150 kHz; a) 0.1% C doubled to 0.2% C; b) 1.0% C doubled to 2.0% C.

From Figure 4-1, the optimal conditions to minimize the operational cost of the acoustic apparatus can be determined. In general, the lower the acoustic intensities and the higher the flow speed, the lower is the operational cost of the acoustic separator.

Several key improvements in the operational cost of the acoustic separator are expected to be seen, which would reduce the power consumption seen in Figure 4-1 significantly:

- Move to a higher frequency, such as 1.5 MHz. Preliminary results using a set of 1.5 MHz transducers in the laboratory acoustic separator at IPST have shown that
between 2 and 5 times less power is required at 1.5 MHz to see the same deflection compared to 150 kHz. This would result in direct energy savings of 50-80%.

- Use single frequency amplifiers instead of broadband amplifiers. Currently, the laboratory acoustic separator uses commercially available broadband RF amplifiers. Due to their broadband nature, these amplifiers have low efficiency (estimated around 50%). The low efficiency shows up as power at harmonics and subharmonics of the transducer driving frequency. The transducer is very narrow band, and hence the power at the harmonics and subharmonics is not used (it is reflected or turned into heat). Switching to a higher efficiency single frequency amplifier (such as a Class D amplifier) will improve energy efficiency to between 75 and 90%.

- Drive transducers in a pulse mode rather than in a continuous mode. Observations have shown that when transducers are operated in a pulse mode, the acoustic force acting on fibers may be larger (at the onset of excitation). If this is indeed the case (additional testing needed to confirm these observations), less power would be required to drive the transducers (e.g., 50% on, 50% off).

- Collect acoustic power which is not used in the apparatus. The stock in the acoustic separation apparatus does not absorb all of the acoustic power put into the separation apparatus. A portion of this energy goes completely through the stock and is currently absorbed on the back side of the apparatus using a silicon absorber. By replacing the absorber with a receiving transducer, the unused acoustic energy can be converted back into electrical energy that can be recycled back into the system.
From the first two improvements, power consumption is estimated to be reduced from 10% to 33% of the current power consumption.

Figure 4-2 shows the area of the transducer necessary to double the pulp consistency as a function of flow speed and acoustic intensity. As can be seen, higher flow speeds and higher acoustic intensities generally result in smaller transducers.

**Figure 4-2.** Transducer area required to double pulp consistency for a 1000 GPM flow at 150 kHz, as a function of flow speed and acoustic intensity; (a) 0.1% C doubled to 0.2% C; (b) 1% C doubled to 2% C.

Table 4-1 shows the cost of building an acoustic separation cell with an area of 1 m². The 1999 column is based upon recent quotes from the current transducer manufacturer. As the transducers used in the acoustic separation apparatus at this time are custom made, significant savings can be realized by switching to a larger scale production facility once the apparatus designs are finalized. The 2002 column is what we believe is obtainable once production is switched to a larger facility. Table 4-1,
combined with Figure 4-2, allows the calculation of the cost of building an acoustic separation apparatus.

**Table 4-I.** Cost of building an AST system.

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<th>1999</th>
<th>2002 (estimate)</th>
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<tr>
<td>Transducer Fabrication, 1 m²</td>
<td>$60,000</td>
<td>$20,000</td>
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<tr>
<td>Housing</td>
<td>$10,000</td>
<td>$2,000</td>
</tr>
<tr>
<td>RF power source (up to 2 W/cm²)</td>
<td>$40,000</td>
<td>$20,000</td>
</tr>
<tr>
<td>Total cost to build a 1-m² acoustic cell, including power amplifiers and housing.</td>
<td>$110,000</td>
<td>$45,000</td>
</tr>
<tr>
<td>Cost per cm²</td>
<td>$11.00</td>
<td>$4.50</td>
</tr>
</tbody>
</table>

For example, for an acoustic separation apparatus to increase the consistency of 1000 GPM of stock from 0.1 to 0.2%, a transducer with an area of approximately 0.4 m² running at 2 W/cm² and 0.4 m/s could be used. At 1999 costs, this would require an initial investment of $44,000 to build. In 2002, the production cost is estimated to be approximately $18,000. From Figure 4-1 a), this acoustic separator would require approximately 12 kW to operate (reduced to 1-4 kW in 3 years due to increases in efficiency).

An acoustic separator used to increase the consistency of 1000 GPM of stock from 1% to 2% could use a transducer with an area of approximately 2 m², running at 6 W/cm² and 0.4 m/s. At 1999 costs, this would require an initial investment of $380,000 (higher than indicated in Table 4-I to compensate for larger RF power source). In 2002, the production cost is estimated to be approximately $164,000. From Figure 4-1 b), the energy requirements would be approximately 110 kW (reduced to 11-37 kW in 3 years due to increases in efficiency).
Based upon equations 4-6 and 4-7, the transducer area and power consumption scale linearly with the flow rate. A flow rate of 100 GPM would require a transducer $1/10^{th}$ the area and use $1/10^{th}$ the power of the ones described above. It should also be noted that increasing consistency another factor of two would theoretically only require half again as much energy as the first factor of two. For example, to go from 0.1% C to 0.2% C for a 1000 GPM flow required 11 kW. To go from 0.1% C to 0.4% C would require 16.5 kW, and to go from 0.1% C to 0.8% C would require 19 kW.

5. Theoretical/Modeling Research Program (Mee Choi)

The acoustic field in the medium can be described by the following parameters: pressure $p$ (local disturbance of the medium), density $\rho$, and velocity $v$ of the fluid particles. The acoustic field carries the energy density $E_0$ in the propagation path. The time-averaged total energy density $\bar{E}_0$ can be written by $p_{max}^2/2\rho_0c^2$ where $p_{max}$ is the maximum value of the pressure for one cycle, $\rho_0$ is the density of the medium under the equilibrium, and $c$ is the sound velocity in the medium.

We consider the transducer as the source of the sound wave. However, in the real situation, the ultrasonic beam radiated by the extended source should be considered equivalent to a large number of point sources. Therefore, unlike the exponential decay of the intensity, we would see the peaks and dips in the pressure for a distance from the transducer due to diffraction. It was reported by Beissner [1987] that a field distribution of the focused beam from a piston is not the same as a plane wave. We have not taken this into consideration, instead we have treated the source simply as a
traveling wave. Any discrepancy between the experimental and theoretical results may come from this simplified assumption.

5.1 Acoustic Radiation Pressure

The acoustic radiation pressure (ARP) is the time average of the pressure acting on an object in a sound field. In a linear approximation, the force is periodic and when averaged over time, it sums to zero. The radiation pressure comes from using higher order terms. The radiation force becomes significant only when the amplitude is high enough. Using hydrodynamics, the acoustic radiation pressure is equal to the time average momentum flux across the surface of the object. The equation of state accompanying the hydrodynamic equation has the form

\[ p' = \rho c^2 + \frac{1}{2} \left( \frac{\partial}{\partial \rho} c^2 \right) \rho'^2 \]  

(5-1)

where \( p' \), \( \rho' \) are the second order variations of the pressure and density respectively, and \( c \) is the sound velocity.

5.1.1 ARP Acting on a Single Particle

Various theoretical results of the force acting on an object of a particular geometry are available. Forces acting on a sphere, a disk, and a cylinder are given by King [1934] (sphere), King [1935] (disk), Awatani [1953 and 1955] (cylinder), Zhuk [1986] (cylinder), and Hasegawa [1988] (cylinder). All derivations above are done by treating the medium as ideal. As a first approximation, a wood pulp fiber can be considered as a rigid cylinder.
A review of the different derivations for the acoustic force applied to rigid and elastic cylinders was undertaken. Derivations by Awatani [1955] and Zhuk [1986] (infinitely long rigid cylinder in traveling wave field with axis perpendicular to the sound field) and Hasegawa et al. [1988] (infinitely long elastic cylinder in traveling wave field with axis perpendicular to the sound field, including internal reflections and scattering) are compared in Figure 5-1.

![Plot of dimensionless acoustic radiation force vs. ka (also dimensionless) for the case of a traveling wave field as obtained using equations from Awatani [1955], Hasegawa [1969] and Zhuk [1986]. Upper axis is cylinder radius, a, in mm for a 150-kHz acoustic wave in water. Awatani and Zhuk give nearly identical results, hence the overlap.](image)

**Figure 5-1.** Plot of dimensionless acoustic radiation force vs. ka (also dimensionless) for the case of a traveling wave field as obtained using equations from Awatani [1955], Hasegawa [1969] and Zhuk [1986]. Upper axis is cylinder radius, $a$, in mm for a 150-kHz acoustic wave in water. Awatani and Zhuk give nearly identical results, hence the overlap.
In Figure 5-1, the dimensionless force (force per unit length $L$, per unit surface area $S$, and per unit acoustic energy density $E$) is plotted as a function of the parameter $ka$ where $k$ is the wave number and $a$ is the cylinder radius. Assuming water as the suspending medium with a density of 1000 kg/m$^3$ and 150 kHz as the wave frequency, $k$ becomes 628 m$^{-1}$. It can be seen that in the range of $ka$ of interest to us ($ka = 0.01$ to 0.1), the derivations agree very well with each other. From Figure 5-1, increasing $ka$ will cause an increase in the dimensionless force. Increasing $ka$ one order of magnitude (say from 0.01 to 0.1) will cause an increase in the dimensionless force of approximately 2.5 orders of magnitude. This could also be interpreted as: an increase of one order of magnitude in $ka$ will reduce the acoustic energy density ($E$), and therefore electrical power, by approximately 2.5 orders of magnitude. Hence, it appears that working in a $ka$ range of 0.01 is not as efficient as working closer to a $ka$ of 1. The value of $ka$ can be increased by two ways: an increase in the cylinder radius ($a$); or an increase in the acoustic wave frequency (increase in $k$). As the fiber dimensions are already determined ($a$), it would be preferable to increase the transducer frequency ($k$).
Figure 5-2. Comparison between acoustic radiation force for traveling and standing wave field for both a rigid sphere and a rigid cylinder, from Awatani [1955]. The acoustic force for a traveling wave field is significantly larger in both cases.

Figure 5-2 shows a comparison of the force between a traveling wave and a standing wave field for both a rigid sphere and a rigid cylinder, as derived by Awatani [1955]. For a nominal fiber of 25-mm radius in water exposed to a 150-kHz sound field ($ka = 0.0157$), a standing wave will produce a force approximately 5000 times greater than the traveling wave. Figure 5-3 plots the ratio between the standing wave force and the traveling wave force. As $ka$ approaches 1, the traveling wave and standing wave forces converge. For low values of $ka$, the standing wave will produce a significantly stronger force than the traveling wave.
Figure 5-3. Ratio of Standing Wave acoustic force to Traveling Wave acoustic force, based on Awatani [1955]. For a fiber with a 25-mm radius at 150 kHz, the ratio is approximately 5000:1.

5.1.2 ARP Acting on an Acoustic Absorber

The acoustic radiation pressure acting on an absorbing object can also be used to deduce the total acoustic power. When an ultrasonic beam travels through a liquid medium and an absorbing material intercepts the entire beam, the Langevin radiation pressure can be used to evaluate the total acoustic power [Rooney and Nyborg, 1972]. The experimental setup for total power measurements was previously described in Section 3 and the apparatus was shown in Figure 3-6.
From the relationship

\[ F = \frac{W}{c} \]  

(5-2)

where \( F \) is the force acting on the absorber (it can be divided by the gravitational constant \( g \) to get an equivalent mass), \( W \) is the acoustic power, and \( c \) is the sound velocity through the medium. As the power goes up, we will expect the force measured to go up proportionally. We, therefore, determine the acoustic pressure from the force \( F \). The acoustic radiation pressure can also be related to the energy density in a perfect absorbing medium [Hamilton and Blackstock, 1998]. The force acting on an absorber in terms of the energy density of the sound wave, i.e., \( E_o \), is

\[ F = 2E_o \]  

(5-3)

This can be utilized for measuring the acoustic field intensity.

The absorber generally spans the entire cross section of the field, and, therefore, the radiation force acting on the absorber is independent of the absorber dimensions.

5.1.3 Measurement of Local ARP and Total Power Associated with ARP

A hydrophone is used to measure the local acoustic radiation pressure. To measure the total power, we set up an imaginary plane parallel to the transducer. We then divide this plane into small segments (i.e., a grid) and measure the local pressure in each of the small segments. The total power associated with ARP (\( W \)) is then obtained.
where $P_{rms}$ is the RMS value of the reading of the pressure, $dA$ is the area of a small segment, and the total number of segments is $N$. The upper bound of the total power associated with ARP is limited to the electric power. We expect a linear relationship between the total acoustic power and the electric power within a reasonable range. However, at large electrical powers, nonlinear effects such as cavitation and acoustic streaming may occur.

5.2 Acoustic Streaming

It has been conjectured that there are other phenomena along with the acoustic radiation pressure such as acoustic streaming (the flow motion of the fluid due to gradual absorption of the sound field), cavitation (bubble activity in a high intensity sound field), and the interrelationships of all of the above.

5.2.1 Calculation for the Acoustic Streaming Based on Nyborg [1965]

Theory of acoustic streaming is based on hydrodynamic equations for viscous compressible fluid. We only consider the motion from a traveling wave in an unbounded medium here. When an ultrasound beam travels in any bounded medium, the presence of the boundaries may affect the streaming. Nyborg [1965] gives a comprehensive literature survey on acoustic streaming. Streaming is reported to occur due to the attenuated effect of the wave as it travels through the medium. His
calculation was based on the approximation of the hydrodynamic equations up to the second order and did not consider nonlinear propagation.

The first-order velocity of the traveling wave, $u_1$, is written as,

$$u_1 = u_0 e^{-(\alpha + ik)x}$$  \hspace{1cm} (5-5)

where $u_0$ is the unattenuated acoustic velocity, $\alpha$ is the attenuation coefficient and $k$ is the wave number. The time-averaged force per unit volume responsible for the flow is written as

$$F_x = \alpha pu_0^2 e^{-2\alpha x}$$  \hspace{1cm} (5-6)

and can be rewritten using the power $W$,

$$F_x = \frac{2\alpha W}{\pi d^2 c}$$  \hspace{1cm} (5-7)

where $d$ is the dimension of the transducer. The velocity $u_2$ for one dimensional axial streaming at position $x$ is given by

$$u_2 = -\frac{\Delta W}{4\pi \mu c d}$$  \hspace{1cm} (5-8)

$$\Delta W = 2\alpha W(x) dx$$  \hspace{1cm} (5-9)

where $\Delta W$ is the energy lost in the acoustic wave while travelling a distance $dx$. The flow direction inside the acoustic beam will be in the positive direction. Conservation of mass requires that there be a flow in the negative direction outside of the acoustic beam.
beam. We can obtain the distance from the acoustic beam at which the flow changes direction from positive to negative. This depends on the ratio of the radius of the source and that of the bounding medium.

5.2.2 Calculation of Acoustic Streaming Based on Eckart [1948]

According to Eckart [1948], three types of streaming are known. The first type is streaming in a viscous boundary layer near obstacles, and the second type is streaming outside the boundary. Both of these have vortex structures. The third type of streaming occurs in a free, non-uniform sound field where the streaming scale is larger than the acoustic wavelength. Eckart [1948] obtained a stationary solution for a long acoustic beam with a radius of $r$. The typical streaming velocity is

$$U = \frac{b}{4\eta c} v_o^2 (kr)^2$$  \hspace{1cm} (5-10)

where $b = 4/3 \eta + \eta'$ (where $\eta$ is the shear viscosity and $\eta'$ is the bulk viscosity), $k$ is the wave number, and $v_o$ is the acoustic velocity. This equation shows that the flow motion arises due to both types of viscosity (shear and bulk).

5.3 Cavitation

No systematic theories have been developed describing what effect cavitation bubbles have on solid objects under ultrasonics, since it would be a very complicated interaction involving three phase media. The bubbles are known to be created when the acoustic pressure amplitude is high enough (so that the time varying amplitude creates pockets of negative pressure) to break the tensile strength of the liquid. Under this condition, the bubbles grow due to the surrounding dissolved gas. This behavior yields interesting
phenomena, such as sonoluminescence, cavitation erosion, chemical reactions, etc. [Apfel 1989, Neppiras 1980]

The effect of cavitation on the acoustic radiation pressure is not well understood. We currently rely on experimental observations of the phenomena. Usually the sound intensity decreases when the acoustic energy is used to create bubbles in a cavitation zone. Therefore, we would expect to observe Eckart type fluid motion.

Some interesting results by Prosperetti [1984] show that the insertion of air bubbles into the medium under ultrasonics can significantly increase the streaming effect. The exact mechanism of this phenomenon is not well understood. Analytical results on cavitation bubbles are currently limited to the study of a single bubble, while in a realistic situation, many bubbles are found in each stream.

5.4 Total Acoustic Force

The total force acting on any object in an acoustic field is the sum of the force due to ARP, streaming and cavitation bubbles. At this time, the force due only to ARP and streaming is considered on two different objects (i.e., cavitation is neglected).

5.4.1 Total Force Acting on an Absorber

If we assume that the sum of the energy density of the sound field and the flow kinetic energy is constant, the average force acting on an absorber can be written as

$$F = F_i + F_s = \frac{W}{c} \tag{5-11}$$
where $F_t$ is the force due to the acoustic radiation pressure, $F_s$ is the force due to the streaming, and $W$ is the equivalent power of the sound energy. We do not include the energy which may be lost due to the formation of cavitation bubbles. Our result from the balance experiment (Section 3) shows discrepancies at different frequencies, as we vary the intensity of the power. The reason is probably due to cavitation bubble losses in the medium.

5.4.2 Total Force Acting on a Fiber Mat

The force acting on a porous medium under ultrasonics would be very hard to calculate, since the frequency dependence of scattered sound waves can generally depend on the internal structures of the porous medium. Also, since the shapes are random, a closed form solution to the acoustic radiation force is difficult to obtain. We can treat the fiber mat as an absorbing object and consider the force acting on the surface. We can use Equation 5-11 to get the upper bounds of the force.

Based on observations, the total force acting on the fiber mat is larger than this estimate, possibly due to the strong water motion which is intensified due to the bubble activities. A speculation would be that when the bubbles collapse, they induce fluid motion which is similar to Eckart [1984] type streaming. This water motion can even be comprised of layers of turbulent jets. For a simplified picture, if the flow velocity is $v_f$, the additional force acting on the surface of the mat will be

$$F_{add} = \frac{1}{2} C_d v_f^2 \rho A \quad (5-12)$$

where $C_d$ is the drag coefficient for the surface of the mat and $A$ is the area of the porous medium.
6. Separation of Vessel Elements and Hardwood Fibers (Feler Bose and Brian Pufahl)

6.1 Introduction

Hardwood fibers are utilized in printing and writing papers to provide a smooth surface with enhanced mechanical properties. However, hardwood stock contains a large number of vessel elements, which can be removed from the surface of the paper during printing. This removal, known as vessel picking, causes a print defect at the point of vessel removal as well as a repetitive print defect where the vessel adheres to the print blanket [Shallhorn and Heintze, 1994]. At this time, no single commercial method has been developed to sufficiently prevent or mask the picking phenomenon, although several have been tried including market-pulp drying [McGovern, 1977] and the use of hydrocyclones [Ohsawa et al., 1984]. For this reason, the most effective method to reduce picking is to monitor the number of vessel elements present in stock and adjust the grade being produced accordingly.

Since there are currently no satisfactory methods to remove vessel elements, the industry addresses the problem by using pulp mixtures to reduce the concentration of vessel elements. Also, refining is used to reduce the size of these particles but at the expense of unnecessary hardwood fiber treatment. At the Fall 1996 Paper Physics Project Advisory Committee meeting, it was recommended that an investigation of the use of acoustic radiation pressure principles to separate vessel elements and hardwood fibers be undertaken.
A preliminary study of acoustic separation of vessels and hardwood fibers was performed by Michelle Oakland (M.S., June 1997) during FY 1996-1997 [Oakland, 1997]. Eucalyptus bleached kraft pulp (100%) from dry lap sheets was used for all experiments. As no standard test method was available for counting vessels within a pulp, a considerable amount of time was redirected to the development of a fast and reliable test method to quantify the number and size of vessels. After several improvements over current test practices in the industry, the test method developed by Michelle Oakland remained inadequate and prevented her from making significant progress in the vessel/hardwood fiber separation method. It was made clear that additional research work needed to be dedicated to the development of a more advanced test method.

Section 6.2 reports on the details of a semi-automated image analysis-based method for vessel size distribution. This method was first reported in March 1998. Significant improvements were made during the past year. Section 6.3 describes the design of a series of acoustic separation experiments to demonstrate the concept of vessel/hardwood fiber separation. Section 6.4 presents results as well as a discussion.

6.2 **Automated Detection of Vessels in Hardwood Furnish (Brian Pufahl)**

6.2.1 **Introduction**

A new method of quantifying hardwood pulp vessel content is presented in this section. This method relies on a CCD imager in conjunction with custom software to determine size distribution of vessels. Operator interaction is limited to the preparation of samples and placement of slides under the imager. This method has been found to be very
robust and provides high accuracy while greatly reducing the analysis time over
traditional measurement methods.

6.2.2 Sample Preparation and Imaging

This system relies on batch-mode processing and requires some preprocessing of the
stock solution. During preprocessing, the consistency of the stock sample is measured
and adjusted to 0.1% (0.015% for lower consistency stock). The stock is then pipetted
with 2.5 ml or 2.5 mg (5 ml of lower consistency stock or 0.75 mg) placed on each of 6
4 x 5-cm slides (providing an order of magnitude increase in area over the traditional
slide). A 2 mm opening on the end of the pipette is critical to prevent large objects from
plugging the opening. The samples are then dried on a hot plate and dyed with Victoria
Blue to enhance optical contrast. Without the staining step, the vessels tend to adhere
to the slide during drying and provide very little contrast. The slides are then provided
with a cover slip to enhance optical transmission and to prevent foreign material from
interfering with the measurements.

The slide is transferred to an automated XY stage mounted below a digital CCD camera
with 10-bit grayscale accuracy. The use of a fully digital CCD camera provides an
artifact-free image that requires much less pre-processing of the image than standard
analog cameras. All camera settings and table positioning commands are controlled by
a single Intel Pentium-based computer (see Figure 6-1). A total of 30 images with a
spatial resolution of 9.05 x 9.05 microns (square) are taken of each slide. A manually
adjusted 5 x 5-cm fiber optic backlight provides transmitted light for the detection of
objects while a fluorescent ring light enhances surface details of detected objects. To
ensure proper size measurements throughout the camera’s field of view, a telecentric
lens system is used. This type of lens virtually eliminates any distortion that would typically be present at the corners of an image.

The LabVIEW-based software system consists of a graphical user interface, routines for controlling the camera and XY stage, routines for processing the images, and a reporting system that includes spreadsheet output of the processed data. Included in the routines for processing the images are algorithms for enhancing the detail of the original image and finding objects exceeding a specified size threshold. The total time to image and analyze a slide is 4 minutes.

The software was "tuned" to look for vessels that would traditionally have been labeled "small to large vessels". White oak stock also contains a number of smaller vessels that do not seem to have a tendency to pick during printing and, therefore, are not measured. The term "equivalent radius" is defined as the radius of a disk having the same surface area as the vessel. By analyzing the subjective values typically used in microscopy, oak vessels are considered significant if they have an equivalent radius above 0.125 mm. Figure 6-2 shows the vessels that are analyzed using both the automated system and the manual microscopy system.
6.3 Vessel/Hardwood Fiber Separation Experiments

As of January 1998, pure white oak pulp (*Quercus alba*) supplied by Champion International was used instead of eucalyptus pulp for the experimental work. White oak
was substituted because eucalyptus is not used very much in the North American
continent to make paper.

The following variables have been considered in the design of separation experiments. They are: pulp consistency, acoustic intensity, flow rate, frequency, acoustic cell width, and mechanical divider position with respect to the transducers. The pulp consistency was run at 0.2 and 0.03%. The acoustic intensity was varied from 0 to 10 W/cm². The flow rate was adjusted from 0.033 to 0.4 m/s. Three frequencies were used: 60 kHz, 150 kHz, and 1.5 MHz. The acoustic cell width (separation distance between transducers and absorbers) was initially set to 5 cm, but was changed to 15 cm on the basis that it would be easier to distinguish vessel-depleted (transducer side) and vessel-enriched (absorber side) output streams. The mechanical divider blade was set at two positions with respect to the transducer side: at the halfway mark (50:50) and also at the one-quarter marks (25:75). Tables 6-1 through 6-3 show the runs that were performed.

Table 6-1. Runs done for 150 kHz and 60 kHz. The consistency was ~0.2% and the divider location was at 50:50.

<table>
<thead>
<tr>
<th>Intensity</th>
<th>0 W/cm²</th>
<th>2 W/cm²</th>
<th>6 W/cm²</th>
<th>10 W/cm²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.033 m/s</td>
<td>X</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.2 m/s</td>
<td>X</td>
<td>X</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.3 m/s</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>0.4 m/s</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
</tbody>
</table>
Table 6-2. Runs done for 1.5 MHz. The shaded region was run at both 0.2% and 0.03% consistency, while the unshaded region was run only at 0.2%. The divider location was at 50:50.

<table>
<thead>
<tr>
<th>Flow (m/s)</th>
<th>Intensity (W/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
</tr>
<tr>
<td>0.033</td>
<td>X</td>
</tr>
<tr>
<td>0.2</td>
<td>X</td>
</tr>
<tr>
<td>0.3</td>
<td>X</td>
</tr>
<tr>
<td>0.4</td>
<td>X</td>
</tr>
</tbody>
</table>

Table 6-3. Runs done for 1.5 MHz. Runs were done at -0.2% consistency. The shaded region were runs done where the divider was at both 25:75 (3.5 cm from transducer) and 50:50, and the unshaded runs were done with a 50:50 divider position.

<table>
<thead>
<tr>
<th>Flow (m/s)</th>
<th>Intensity (W/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
</tr>
<tr>
<td>0.033</td>
<td>X</td>
</tr>
<tr>
<td>0.2</td>
<td>X</td>
</tr>
<tr>
<td>0.3</td>
<td>X</td>
</tr>
<tr>
<td>0.4</td>
<td>X</td>
</tr>
</tbody>
</table>

6.4 Results (Feler Bose)

This section describes the results of the runs previously illustrated in Tables 6-1 to 6-3. All results are shown in tabular form. Selected results are shown in graphical form.

There are five tables in this section, each corresponding to a different set of experiments. For each table:

- First column is acoustic intensity (applied electrical power divided by the area of the transducer)
- Second column is flow velocity
- Third and fourth columns are the total number of vessels (for six slides) as measured in the absorber and transducer output streams, respectively, using the test method described in Section 6.2

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Fifth column is the total number of vessels for the six slides (combined absorber and transducer streams)
Sixth column is the difference of vessels between absorber and transducer streams
Seventh column is the difference of vessels divided by the total number of vessels in percentage.

An error analysis is also presented for each data set. The consistency measurement error is in agreement with TAPPI T-240 (Consistency (concentration) of pulp suspensions). The adjustment to 0.1% (or 0.015%) consistency is based on the fact that there will be an error when one adjusts the consistency in various beakers. The pipette error is based on reading error of the pipette of 0.05 ml. The vessel counting error is based on the image analysis system. This error is due to the fact that the camera does not count vessels that appear at the edge of the image. Total error is the sum of all the errors.
Table 6-4. Results of 1.5-MHz transducer, ~0.2% consistency, and divider at 50:50.

<table>
<thead>
<tr>
<th>Intensity (W/cm²)</th>
<th>Velocity (m/s)</th>
<th>Absorber Stream # of Vessels</th>
<th>Transducer Stream # of Vessels</th>
<th>Total</th>
<th>Difference</th>
<th>% of Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.033</td>
<td>890</td>
<td>1134</td>
<td>2024</td>
<td>-244</td>
<td>-12.06%</td>
</tr>
<tr>
<td>1</td>
<td>0.2</td>
<td>1074</td>
<td>1274</td>
<td>2348</td>
<td>-200</td>
<td>-8.52%</td>
</tr>
<tr>
<td>1</td>
<td>0.3</td>
<td>940</td>
<td>1087</td>
<td>2027</td>
<td>-147</td>
<td>-7.25%</td>
</tr>
<tr>
<td>1</td>
<td>0.4</td>
<td>1197</td>
<td>877</td>
<td>2074</td>
<td>320</td>
<td>15.43%</td>
</tr>
<tr>
<td>3</td>
<td>0.2</td>
<td>1022</td>
<td>1085</td>
<td>2107</td>
<td>-63</td>
<td>-2.99%</td>
</tr>
<tr>
<td>3</td>
<td>0.3</td>
<td>908</td>
<td>1080</td>
<td>1988</td>
<td>-172</td>
<td>-8.65%</td>
</tr>
<tr>
<td>3</td>
<td>0.4</td>
<td>1025</td>
<td>1258</td>
<td>2283</td>
<td>-233</td>
<td>-10.21%</td>
</tr>
<tr>
<td>0</td>
<td>0.2</td>
<td>1475</td>
<td>855</td>
<td>2330</td>
<td>620</td>
<td>26.61%</td>
</tr>
<tr>
<td>1</td>
<td>0.2</td>
<td>1074</td>
<td>1274</td>
<td>2348</td>
<td>-200</td>
<td>-8.52%</td>
</tr>
<tr>
<td>2</td>
<td>0.2</td>
<td>1047</td>
<td>1158</td>
<td>2205</td>
<td>-111</td>
<td>-5.03%</td>
</tr>
<tr>
<td>3</td>
<td>0.2</td>
<td>1022</td>
<td>1085</td>
<td>2107</td>
<td>-63</td>
<td>-2.99%</td>
</tr>
<tr>
<td>0</td>
<td>0.3</td>
<td>1153</td>
<td>1128</td>
<td>2281</td>
<td>25</td>
<td>1.10%</td>
</tr>
<tr>
<td>1</td>
<td>0.3</td>
<td>940</td>
<td>1087</td>
<td>2027</td>
<td>-147</td>
<td>-7.25%</td>
</tr>
<tr>
<td>2</td>
<td>0.3</td>
<td>908</td>
<td>1080</td>
<td>1988</td>
<td>-172</td>
<td>-8.65%</td>
</tr>
<tr>
<td>0</td>
<td>0.4</td>
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<td>1227</td>
<td>2603</td>
<td>149</td>
<td>5.72%</td>
</tr>
<tr>
<td>1</td>
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<td>1197</td>
<td>877</td>
<td>2074</td>
<td>320</td>
<td>15.43%</td>
</tr>
<tr>
<td>2</td>
<td>0.4</td>
<td>1159</td>
<td>1323</td>
<td>2482</td>
<td>-164</td>
<td>-6.61%</td>
</tr>
<tr>
<td>3</td>
<td>0.4</td>
<td>1025</td>
<td>1258</td>
<td>2283</td>
<td>-233</td>
<td>-10.21%</td>
</tr>
</tbody>
</table>

Mean Number of Vessels 2195
Standard Deviation 194 8.84%
Range 1988 2603
Range as Percentage of Mean -9.42% 18.60%

**Error Sources (+)**

<table>
<thead>
<tr>
<th>Source</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measurement of Consistency</td>
<td>30 per 6 slides</td>
</tr>
<tr>
<td>Adjustment to 0.1% Consistency</td>
<td>11 per 6 slides</td>
</tr>
<tr>
<td>Pipette Error</td>
<td>4 per 6 slides</td>
</tr>
<tr>
<td>Vessel Counting Error</td>
<td>33 per 6 slides</td>
</tr>
<tr>
<td>Total Error</td>
<td>78 per 6 slides</td>
</tr>
<tr>
<td>Percentage Error</td>
<td>7.13% per 6 slides</td>
</tr>
</tbody>
</table>

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Table 6-5. Results of 1.5-MHz transducer, ~0.03% consistency, and divider at 50:50.

<table>
<thead>
<tr>
<th>Intensity (W/cm²)</th>
<th>Velocity (m/s)</th>
<th>Absorber Stream # of Vessels</th>
<th>Transducer Stream # of Vessels</th>
<th>Total</th>
<th>Difference</th>
<th>% of Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.2</td>
<td>271</td>
<td>292</td>
<td>563</td>
<td>-21</td>
<td>-3.73%</td>
</tr>
<tr>
<td>1</td>
<td>0.3</td>
<td>181</td>
<td>260</td>
<td>441</td>
<td>-79</td>
<td>-17.91%</td>
</tr>
<tr>
<td>1</td>
<td>0.4</td>
<td>343</td>
<td>342</td>
<td>685</td>
<td>1</td>
<td>0.15%</td>
</tr>
<tr>
<td>2</td>
<td>0.2</td>
<td>327</td>
<td>115</td>
<td>442</td>
<td>212</td>
<td>47.96%</td>
</tr>
<tr>
<td>2</td>
<td>0.3</td>
<td>317</td>
<td>235</td>
<td>552</td>
<td>82</td>
<td>14.86%</td>
</tr>
<tr>
<td>2</td>
<td>0.4</td>
<td>121</td>
<td>252</td>
<td>373</td>
<td>-131</td>
<td>-35.12%</td>
</tr>
<tr>
<td>0</td>
<td>0.2</td>
<td>191</td>
<td>264</td>
<td>455</td>
<td>-73</td>
<td>-16.04%</td>
</tr>
<tr>
<td>1</td>
<td>0.2</td>
<td>271</td>
<td>292</td>
<td>563</td>
<td>-21</td>
<td>-3.73%</td>
</tr>
<tr>
<td>2</td>
<td>0.2</td>
<td>327</td>
<td>115</td>
<td>442</td>
<td>212</td>
<td>47.96%</td>
</tr>
<tr>
<td>0</td>
<td>0.3</td>
<td>301</td>
<td>337</td>
<td>638</td>
<td>-36</td>
<td>-5.64%</td>
</tr>
<tr>
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<td>0.3</td>
<td>181</td>
<td>260</td>
<td>441</td>
<td>-79</td>
<td>-17.91%</td>
</tr>
<tr>
<td>2</td>
<td>0.3</td>
<td>317</td>
<td>235</td>
<td>552</td>
<td>82</td>
<td>14.86%</td>
</tr>
<tr>
<td>0</td>
<td>0.4</td>
<td>258</td>
<td>259</td>
<td>517</td>
<td>-1</td>
<td>-0.19%</td>
</tr>
<tr>
<td>1</td>
<td>0.4</td>
<td>343</td>
<td>342</td>
<td>685</td>
<td>1</td>
<td>0.15%</td>
</tr>
<tr>
<td>2</td>
<td>0.4</td>
<td>121</td>
<td>252</td>
<td>373</td>
<td>-131</td>
<td>-35.12%</td>
</tr>
</tbody>
</table>

Mean Number of Vessels: 518
Standard Deviation: 101 (19.82%)
Range: 373-685 (61.14%)
Range as Percentage of Mean: -28.05% 32.13%

Error Sources (±):
- Measurement of Consistency: 46 per 6 slides
- Adjustment to 0.015% Consistency: 3 per 6 slides
- Pipette Error: 1 per 6 slides
- Vessel Counting Error: 16 per 6 slides
- Total Error: 66 per 6 slides
- Percentage Error: 25.40% per 6 slides
Table 6-6. Results of 1.5-MHz transducer, ~0.2% consistency, and divider at 25:75 (25% of feed stream to transducer stream).

<table>
<thead>
<tr>
<th>Intensity (W/cm²)</th>
<th>Velocity (m/s)</th>
<th>Absorber Stream # of Vessels</th>
<th>Transducer Stream # of Vessels</th>
<th>Total</th>
<th>Difference</th>
<th>% of Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.2</td>
<td>958</td>
<td>984</td>
<td>1942</td>
<td>-26</td>
<td>-1.34%</td>
</tr>
<tr>
<td>0</td>
<td>0.4</td>
<td>918</td>
<td>999</td>
<td>1917</td>
<td>-81</td>
<td>-4.23%</td>
</tr>
<tr>
<td>2</td>
<td>0.2</td>
<td>995</td>
<td>1162</td>
<td>2157</td>
<td>-167</td>
<td>-7.74%</td>
</tr>
<tr>
<td>2</td>
<td>0.3</td>
<td>985</td>
<td>981</td>
<td>1966</td>
<td>4</td>
<td>0.20%</td>
</tr>
<tr>
<td>2</td>
<td>0.4</td>
<td>1037</td>
<td>1057</td>
<td>2094</td>
<td>-20</td>
<td>-0.96%</td>
</tr>
</tbody>
</table>

Mean Number of Vessels

<table>
<thead>
<tr>
<th>Mean Number of Vessels</th>
<th>2015</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard Deviation</td>
<td>105</td>
</tr>
<tr>
<td>Range</td>
<td>1917</td>
</tr>
<tr>
<td>Range as Percentage of Mean</td>
<td>-4.87%</td>
</tr>
</tbody>
</table>

Error Sources (+)

<table>
<thead>
<tr>
<th>Error Sources (+)</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Measurement of Consistency</td>
<td>28 per 6 slides</td>
</tr>
<tr>
<td>Adjustment to 0.1% Consistency</td>
<td>10 per 6 slides</td>
</tr>
<tr>
<td>Pipette Error</td>
<td>3 per 6 slides</td>
</tr>
<tr>
<td>Vessel Counting Error</td>
<td>32 per 6 slides</td>
</tr>
<tr>
<td>Total Error</td>
<td>73 per 6 slides</td>
</tr>
<tr>
<td>Percentage Error</td>
<td>7.26% per 6 slides</td>
</tr>
</tbody>
</table>
Table 6-7. Results of 150-kHz transducer, ~0.2% consistency, and divider at 50:50.

<table>
<thead>
<tr>
<th>Intensity (W/cm²)</th>
<th>Velocity (m/s)</th>
<th>Absorber Stream # of Vessels</th>
<th>Transducer Stream # of Vessels</th>
<th>Total</th>
<th>Difference</th>
<th>% of Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.4</td>
<td>1043</td>
<td>1007</td>
<td>2050</td>
<td>36</td>
<td>1.76%</td>
</tr>
<tr>
<td>2</td>
<td>0.033</td>
<td>1003</td>
<td>1001</td>
<td>2004</td>
<td>2</td>
<td>0.10%</td>
</tr>
<tr>
<td>2</td>
<td>0.3</td>
<td>850</td>
<td>1037</td>
<td>1887</td>
<td>-187</td>
<td>-9.91%</td>
</tr>
<tr>
<td>2</td>
<td>0.4</td>
<td>975</td>
<td>1036</td>
<td>2011</td>
<td>-61</td>
<td>-3.03%</td>
</tr>
<tr>
<td>6</td>
<td>0.2</td>
<td>937</td>
<td>1027</td>
<td>1964</td>
<td>-90</td>
<td>-4.58%</td>
</tr>
<tr>
<td>6</td>
<td>0.3</td>
<td>838</td>
<td>961</td>
<td>1799</td>
<td>-123</td>
<td>-6.84%</td>
</tr>
<tr>
<td>6</td>
<td>0.4</td>
<td>967</td>
<td>793</td>
<td>1760</td>
<td>174</td>
<td>9.89%</td>
</tr>
<tr>
<td>10</td>
<td>0.3</td>
<td>830</td>
<td>901</td>
<td>1731</td>
<td>-71</td>
<td>-4.10%</td>
</tr>
<tr>
<td>10</td>
<td>0.4</td>
<td>958</td>
<td>1077</td>
<td>2035</td>
<td>-119</td>
<td>-5.85%</td>
</tr>
<tr>
<td>2</td>
<td>0.3</td>
<td>850</td>
<td>1037</td>
<td>1887</td>
<td>-187</td>
<td>-9.91%</td>
</tr>
<tr>
<td>6</td>
<td>0.3</td>
<td>838</td>
<td>961</td>
<td>1799</td>
<td>-123</td>
<td>-6.84%</td>
</tr>
<tr>
<td>10</td>
<td>0.3</td>
<td>830</td>
<td>901</td>
<td>1731</td>
<td>-71</td>
<td>-4.10%</td>
</tr>
<tr>
<td>0</td>
<td>0.4</td>
<td>1043</td>
<td>1007</td>
<td>2050</td>
<td>36</td>
<td>1.76%</td>
</tr>
<tr>
<td>2</td>
<td>0.4</td>
<td>975</td>
<td>1036</td>
<td>2011</td>
<td>-61</td>
<td>-3.03%</td>
</tr>
<tr>
<td>6</td>
<td>0.4</td>
<td>967</td>
<td>793</td>
<td>1760</td>
<td>174</td>
<td>9.89%</td>
</tr>
<tr>
<td>10</td>
<td>0.4</td>
<td>958</td>
<td>1077</td>
<td>2035</td>
<td>-119</td>
<td>-5.85%</td>
</tr>
</tbody>
</table>

Mean Number of Vessels: 1916
Standard Deviation: 125
Range: 1731
Range as Percentage of Mean: 6.59%

Error Sources (±):
- Measurement of Consistency: 27 per 6 slides
- Adjustment to 0.1% Consistency: 1 per 6 slides
- Pipette Error: 3 per 6 slides
- Vessel Counting Error: 31 per 6 slides
- Total Error: 62 per 6 slides
- Percentage Error: 6.45% per 6 slides

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Table 6-8. Result of 60-kHz transducer, ~0.2% consistency, and divider at 50:50.

<table>
<thead>
<tr>
<th>Intensity (W/cm²)</th>
<th>Velocity (m/s)</th>
<th>Absorber Stream # of Vessels</th>
<th>Transducer Stream # of Vessels</th>
<th>Total</th>
<th>Difference</th>
<th>% of Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.2</td>
<td>923</td>
<td>959</td>
<td>1882</td>
<td>-36</td>
<td>-1.91%</td>
</tr>
<tr>
<td>0.0</td>
<td>0.4</td>
<td>982</td>
<td>1106</td>
<td>2088</td>
<td>-124</td>
<td>-5.94%</td>
</tr>
<tr>
<td>6.0</td>
<td>0.2</td>
<td>831</td>
<td>810</td>
<td>1641</td>
<td>21</td>
<td>1.28%</td>
</tr>
<tr>
<td>6.0</td>
<td>0.3</td>
<td>946</td>
<td>1001</td>
<td>1947</td>
<td>-55</td>
<td>-2.82%</td>
</tr>
<tr>
<td>6.0</td>
<td>0.4</td>
<td>945</td>
<td>980</td>
<td>1925</td>
<td>-35</td>
<td>-1.82%</td>
</tr>
<tr>
<td>2.0</td>
<td>0.3</td>
<td>905</td>
<td>947</td>
<td>1852</td>
<td>-42</td>
<td>-2.27%</td>
</tr>
<tr>
<td>6.0</td>
<td>0.3</td>
<td>946</td>
<td>1001</td>
<td>1947</td>
<td>-55</td>
<td>-2.82%</td>
</tr>
<tr>
<td>10.0</td>
<td>0.3</td>
<td>1038</td>
<td>969</td>
<td>2007</td>
<td>69</td>
<td>3.44%</td>
</tr>
<tr>
<td>0.0</td>
<td>0.4</td>
<td>982</td>
<td>1106</td>
<td>2088</td>
<td>-124</td>
<td>-5.94%</td>
</tr>
<tr>
<td>2.0</td>
<td>0.4</td>
<td>1135</td>
<td>1098</td>
<td>2233</td>
<td>37</td>
<td>1.66%</td>
</tr>
<tr>
<td>6.0</td>
<td>0.4</td>
<td>945</td>
<td>980</td>
<td>1925</td>
<td>-35</td>
<td>-1.82%</td>
</tr>
<tr>
<td>10.0</td>
<td>0.4</td>
<td>1097</td>
<td>995</td>
<td>2092</td>
<td>102</td>
<td>4.88%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Mean Number of Vessels</th>
<th>1963</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard Deviation</td>
<td>170</td>
</tr>
<tr>
<td>Range</td>
<td>1641</td>
</tr>
<tr>
<td>Range as Percentage of Mean</td>
<td>-16.40%</td>
</tr>
</tbody>
</table>

Error Sources (±)
- Measurement of Consistency: 27 per 6 slides
- Adjustment to 0.1% Consistency: 10 per 6 slides
- Pipette Error: 3 per 6 slides
- Vessel Counting Error: 31 per 6 slides
- Total Error: 72 per 6 slides
- Percentage Error: 7.30% per 6 slides

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Selected results are presented in graphical form in Figures 6-3 to 6-12. The error bars on the graphs are based only on error in total count and do not include error at each equivalent radius. Figure 6-3 shows the best results obtained with the 150 kHz separation trials. A clear concentration of vessels on the vessel-enriched stream (absorber side) can be seen at all vessel sizes.

![Graph of vessel concentration vs. equivalent radius.](image)

**Figure 6-3.** 150 kHz, 0.4 m/s, 0.2%C, 6 W/cm². Divider blade was at 50:50.

Figure 6-4, however, shows results contradicting those found in Figure 6-3. The vessel-depleted stream (transducer side) had a higher concentration at all vessel sizes. The error bars, showing a single standard deviation, are quite close indicating that any potential separation is likely lost in experimental error.
Figure 6-4. 150 kHz, 0.3 m/s, 0.2%C, 2 W/cm². Divider blade was at 50:50.

Figure 6-5 shows typical results run with the 60 kHz transducers. In all runs, the vessel-enriched and vessel-depleted streams have similar amounts of vessels, with the one-sigma error bars clearly overlapping. The complete lack of separation in combination with the marginal results at 150 kHz lead to a potential trend indicating that higher frequencies may improve the separation.
Figure 6-6 shows one of the better runs using the 1.5-MHz transducer. Separation is better than seen at 150 kHz in Figure 6-3. Lower consistency runs were also done at 1.5 MHz to determine the effect of consistency, and Figure 6-7 again shows clear separation.

**Figure 6-6.** 1.5 MHz, 0.4 m/s, 0.2%C, 1 W/cm$^2$. Divider blade was at 50:50.

**Figure 6-7.** 1.5 MHz, 0.2 m/s, 0.03%C, 2 W/cm$^2$. Divider blade was at 50:50.
However, not all of the results were as expected. Figure 6-8 shows 1.5 MHz results, and the vessel-depleted stream shows higher vessel concentrations than the vessel-enriched stream.

![Diagram](image)

**Figure 6-8.** 1.5 MHz, 0.4 m/s, 0.03% C, 2 W/cm². Divider blade was at 50:50.

In an attempt to determine what was going on, samples were collected from the vessel-enriched and vessel-depleted streams at zero power. In many cases (such as Figure 6-9), a statistically similar number of vessels was found in both streams. However, in Figure 6-10, a large discrepancy was found between the vessel-enriched and vessel-depleted streams with no applied power.
Figure 6-9. 1.5 MHz, 0.2 m/s, 0.03%C, 0 W/cm². Divider blade was at 50:50.

Figure 6-10. 1.5 MHz, 0.2 m/s, 0.2%C, 0 W/cm². Divider blade was at 50:50.
Figure 6-11 Results of image analysis on vessel count using no ultrasonic field, Divider blade was at 50:50.

In order to answer some discrepancies observed, experiments were repeated for the cases where no ultrasonic field was used. Figure 6-11 shows the results of those trials. It seems that there could be minor errors associated in the experimental and counting procedures. Nevertheless, the range of errors is within ±5%, which seems reasonable, as opposed to currently available techniques. At the time of writing, a full set of
experiments is repeated using 1.5-MHz transducer to narrow down the possible sources of error.

Further experiments were performed looking at the effect of the divider position. The divider was moved from the middle (50:50) of the flow toward the vessel-enriched side (25:75). Figure 6-12 shows typical results, indicating that separation was not obvious.

![Graph](attachment:image.png)

**Figure 6-12.** 1.5 MHz, 0.2 m/s, 0.2%C, 2 W/cm\(^2\). Divider at 25:75.

**Discussion**

Work done at 60 kHz showed that separation of vessels from fibers does not occur at this frequency. Experiments at 150 kHz showed that, in some cases, statistically significant results could be achieved. However, many of the experiments only showed that any separation was lost in the experimental noise. Based upon the trend of complete lack of separation at 60 kHz, and partial separations at 150 kHz, transducers at 1.5 MHz were ordered and installed. Experiments at 1.5 MHz at times yielded encouraging separations, but an equal number of experiments contradicted these results. Data at zero power yielded results (see Figures 6-9, 6-10 and 6-11) which
indicate that there are some possible errors which entered into the results and which did not occur for trials performed using 60-kHz and 150-kHz transducers. At this time, further experiments to determine the source of the error for measurements obtained using 1.5-MHz transducers are being conducted. Possible explanations, which are being investigated, include:

- Back flow in the flow development section may be leading to a nonuniform velocity profile in the separation section.
- Consistency measurements typically contributed to the largest amount of experimental error (see Tables 6-4 through 6-8).
- Dilution of the sample before pipetting onto the slide may be an unnecessary extra step in the slide preparation.
- Use larger slides than the 4 x 5cm slides to increase the amount of sample measured.
- Try an intermediate frequency, such as 500 kHz. If $ka$ is greater than 1 at 1.5 MHz, then we have passed the maximum efficiency point for the separation.
7. Literature Cited


FUNDAMENTALS OF DIMENSIONAL STABILITY

STATUS REPORT

FOR

PROJECT F020

Douglas Coffin
Barry Hojjatie
Kennisha Collins

Institute of Paper Science and Technology
500 10th Street, N. W.
Atlanta, Georgia 30318
DUES-FUNDED PROJECT SUMMARY

Project Title: Dimensional Stability
Project Code: DIMSTAB
Project Number: F020
PAC: Paper Physics
Division: Fiber and Paper Physics
Project Staff: PI: Douglas Coffin
Staff: Barry Hojjatie, Kennisha Collins
FY 98-99 Budget: $109,564
Time Allocation: Faculty: 25%, Staff: 60%

RESEARCH LINE/ROADMAP:
11 Convertibility and End-Use Performance

Improve the ratio of product performance to cost for pulp and paper products 25% by developing: models, algorithms and functional samples of fibrous structures and coatings which describe and demonstrate improved convertibility and end-use performance.

PROJECT OBJECTIVE:
Reduce the amount of paper rejected because of cockle through improved efficiency in identifying the causes of cockle and/or use of corrective measures to prevent cockle.

To develop a science-based understanding of the dimensional stability of paper and paperboard, especially the phenomenon of cockle, and to apply these fundamental results to practical industrial problems.
PROJECT BACKGROUND

This project was initiated in July of 1994. The scope of work for this project is to gain an understanding of cockle in paper and to develop the knowledge and tools required to eliminate its occurrence. Cockle is a manifestation of the dimensional instability of paper due to local variations in the physical state of the paper coupled with a change in moisture content. The phenomenon of cockle is directly related to the mechanical, hygroexpansive, and physical properties of paper. These properties will be a result of the constituent materials and the papermaking process used to produce the sheet. With an understanding of how cockle occurs, steps can be taken to eliminate it by modifying the constituent materials or papermaking process. Since cockle is inherently a complex problem and is influenced by many different factors, the advances made in this research program will benefit the scientific understanding in all areas of dimensional stability.

SUMMARY OF RESULTS (March 98-March 99)

- Developed methods to produce cockled handsheets for which the cockle resembles cockle in machine-made paper.

- Conducted handsheet study of cockle severity versus papermaking.

- Developed methods to measure and quantify cockle.

- Developed equations to model cockle (moved to student work).

• Presentation on cockle mechanics made at International Paper Physics Conference.

PROJECT GOALS FOR FY 99-00

• Conduct handsheet study to determine the effect of papermaking parameters on cockle that develops after initially flat and dry sheets are exposed to high humidity.

• Conduct handsheet study to better characterize the effect of papermaking parameters on cockle that develops during drying.

• Summarize results and prepare final project report.

PROJECT DELIVERABLES: Report giving relationships between papermaking parameters and severity of cockle. Methods to evaluate cockle, Strategies to eliminate cockle.

PROJECT SCHEDULE

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<th>98b</th>
<th>99a</th>
<th>99b</th>
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</table>
DISCUSSION

The dimensional stability of paper is one of the primary concerns for many paper grades. The interaction between moisture and paper affects the entire life of paper from forming, through converting, to the end-use of the final product. Dimensional instabilities, such as curl and cockle, are blatant examples of the detrimental aspects of moisture and paper. Not so obvious are the loss of modulus or the accelerated creep caused by the interaction of moisture and paper. Thus, gains in the fundamental understanding of dimensional stability are essential to improving the efficiency of the papermaking process.

Cockle is one such area that has not been sufficiently addressed in a scientific manner yet is prevalent in the paper industry. Thus, cockle was chosen as the focus of our excursion into dimensional instability. The plan for the current project was based on the following hypothesis for the cause of cockle.

Cockle results from an in-plane variation of the free hygroexpansive strains resulting from a change in moisture content of the sheet. The nonuniformity in the potential for free expansion creates compressive stresses in the sheet. If the compressive stresses are of sufficient magnitude, the sheet will buckle out-of-plane, thus creating cockle. The free hygroexpansive strain is the stress-free strain resulting from a change in dimensions of a material undergoing a change in moisture content. In addition to the nonuniformity in the free hygroexpansive strains, planar nonuniformities in mechanical or physical properties must be present to induce local buckling of the sheet.

In trying to get to our destination, minimizing cockle, we have set out on three different roadways. Along each roadway, we hope to gather sufficient evidence so
that at the end of our path, we clearly see that we have arrived. The three roadways all head in the same direction and intersect each other along the way.

First, we state that cockle is a buckling phenomenon, and so we are studying the theory of plate buckling. Second, we believe that buckling arises from the restrained expansion/shrinkage of the sheet; consequently, we are investigating hygrobuckling of a sheet. Third, we propose that cockling arises from planar variations in properties arising from variability introduced in papermaking; thus, we are studying the effects of variability and process conditions on the severity of cockle. The following highlights our progress made in the last year on each of these avenues of research.

**Buckling and Postbuckling Theory**

We formulated a model of sheet buckling that should predict cockle under a certain set of parameters. The model accounts for planar variations in thickness, mechanical properties, and moisture histories. The sheet is modeled as a layered structure to account for two-sidedness. Imperfections in the original flatness of the sheet are taken into account. Initially, we have treated the sheet as a thin elastic material but may need to account for inelastic effects. We have assigned orthotropic material properties that may vary both in magnitude and orientation from location to location within each layer of the sheet. For this idealized material, which we hope captures the essence of paper, we will predict the deformed geometry of the sheet when it is subjected to a change in moisture content. In other words, we will try to simulate cockle.

It is probably not possible to predict the exact buckled shape for a specific piece of paper, but we expect to predict the general trends of cockle as a function of the type
and degree of variability. Thus, the model can be used as a tool to study sensitivities and determine the most likely causes of cockle.

The report that presents this formulation is being prepared as a technical report. During the spring meeting last year, the Paper Physics PAC requested that we minimize our effort on the modeling aspects and focus on the experimental program. We accommodated these wishes and did not implement our formulation to obtain numerical results. We have moved this aspect of the work to student research projects. The first-year M.S. student who had begun work in this area, has withdrawn from IPST. We hope to interest another student in the next year.

**Hygroexpansive Buckling**

In order to demonstrate that moisture-induced buckling of paper occurs and is a predictable phenomenon, we studied the hygrobuckling of paper strips as previously reported in last year's project report. In that report, we successfully compared the experimental results to results from an analytic solution of the problem. This year our analytic solution was accepted as a paper by the *International Journal of Nonlinear Mechanics*. We derived an analytic solution that consists of two coupled elliptic integrals. This solution followed the method of Euler, who first solved the elastica problem in the early 1700's. The elastica solution is valid for an inextensible rod subjected to an applied compressive thrust. The difference between Euler's solution and the present solution is that the present solution accounts for the extensibility of the material and the driving force that leads to buckling is due to restrained swelling not an applied load. This solution yields a non-trivial solution in the limit as the bending stiffness goes to zero. This limit corresponds to the case of a string.
Handsheet Study

During this past year, the majority of our effort was spent on conducting a handsheet study of cockle. We wanted to study the effects of different paper-making variables on the severity of cockle. In order to accomplish this task, we first had to develop a method to produce handsheets that were cockled. Once this was accomplished, we chose parameters that we expected would impact cockle and produced a set of sheets to determine the sensitivity of the variables on cockle severity. The results of this study are presented below.

Our goal was to make handsheets with cockle that resembled cockle in machine-made paper. We produced sheets using different techniques of forming and drying until we found a reproducible method to form sheet cockle. It was important to us that we did not deliberately induce the cockle but form it as a consequence of the papermaking process. It is ironic that the papermaker desires to make cockle-free paper and we desire to make cockled sheets, yet both goals are difficult to achieve. This difference in itself may add insight into cockle.

We found that the right conditions during drying lead to cockled sheets. If a handsheet is freely dried, it forms large wrinkles and may even close in upon itself. If a sheet is dried under full restraint, it will be flat. We observed this even if the sheet had nonuniformities. If the sheet is dried under partial restraint, it may lead to cockle. We found that a sheet dried under partial restraint at room temperature tended to form large waves rather than cockle. If we dried the sheet faster, such as on a hot metal plate, we found that the sheet tended to form cockle.
The end result of our drying tests was a method to produce cockled sheets. We formed sheets in a Noble and Wood handsheet mold. This produced an 8 in x 8 in sheet. We modified the formation of the sheets by using a slurry at higher consistency, and delaying the time lapsed before the water was drained. The formed sheets were placed between two blotters and pressed. The pressing pressure varied. The sheet was then dried on an Emmerson Plate Dryer under different conditions. The temperature of the dryer was varied, and the sheet was placed between either 0, 1, or 2 blotters. This method of drying produced cockle to different degrees of severity. Figure 1 shows an example of the type of cockle that was produced in the sheets.

![Figure 1. Example of cockle in handsheets.](image)

Once we developed a method to produce cockled handsheets, we conducted a study to determine the effects of various papermaking parameters on cockle. We
used an 8-test 2-level Plackett-Burman design to prescribe the conditions under which the sheets were produced. In this test, high and low levels of each variable are chosen, then prescribed combinations of the high and low variables, for up to 7 variables, are used to make 8 different sheets. The sheet topography is measured with the shadow Moiré system. The data is analyzed by first averaging the measurements for the sheets at both the low level and high level for a particular variable and then taking the difference in these two measurements. This difference indicates the sensitivity of the measurement to the particular variable. The sheets are made such that the main effect of the other variables has null effect, but interactions between two or more variables may confound the results.

For this study, we chose the following parameters; pulp freeness, grammage, forming, pressing pressure, drying restraint, and drying temperature. Table 1 gives the Plackett-Burman design for the papermaking variables.

Table 1. Plackett-Burman Design for Handsheets

<table>
<thead>
<tr>
<th>Sheet</th>
<th>CSF</th>
<th>Grammage (gsm)</th>
<th>Forming</th>
<th>Pressing (psi)</th>
<th>Restraint (blotters)</th>
<th>Drying Temperature (° C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>350</td>
<td>70</td>
<td>modified</td>
<td>60</td>
<td>0</td>
<td>110</td>
</tr>
<tr>
<td>2</td>
<td>500</td>
<td>90</td>
<td>standard</td>
<td>60</td>
<td>1</td>
<td>110</td>
</tr>
<tr>
<td>3</td>
<td>350</td>
<td>90</td>
<td>standard</td>
<td>0</td>
<td>0</td>
<td>90</td>
</tr>
<tr>
<td>4</td>
<td>350</td>
<td>70</td>
<td>standard</td>
<td>0</td>
<td>1</td>
<td>110</td>
</tr>
<tr>
<td>5</td>
<td>500</td>
<td>70</td>
<td>modified</td>
<td>0</td>
<td>1</td>
<td>90</td>
</tr>
<tr>
<td>6</td>
<td>350</td>
<td>90</td>
<td>modified</td>
<td>60</td>
<td>1</td>
<td>90</td>
</tr>
<tr>
<td>7</td>
<td>500</td>
<td>70</td>
<td>standard</td>
<td>60</td>
<td>0</td>
<td>90</td>
</tr>
<tr>
<td>8</td>
<td>500</td>
<td>90</td>
<td>modified</td>
<td>0</td>
<td>0</td>
<td>110</td>
</tr>
</tbody>
</table>
Three sheets were produced for each of the eight conditions given in Table 1. After drying, the sheets were conditioned at 50% RH. The properties of the sheet are given in Table 2.

<table>
<thead>
<tr>
<th>Sheet</th>
<th>Grammage (gsm)</th>
<th>Hard Platen Density (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>69.7 (2.4)</td>
<td>0.4 (5.9)</td>
</tr>
<tr>
<td>2</td>
<td>92.0 (7.7)</td>
<td>0.5 (0.9)</td>
</tr>
<tr>
<td>3</td>
<td>90.6 (6.7)</td>
<td>0.4 (1.9)</td>
</tr>
<tr>
<td>4</td>
<td>70.0 (2.4)</td>
<td>0.4 (2.3)</td>
</tr>
<tr>
<td>5</td>
<td>72.3 (6.7)</td>
<td>0.3 (2.3)</td>
</tr>
<tr>
<td>6</td>
<td>93.6 (2.7)</td>
<td>0.5 (3.3)</td>
</tr>
<tr>
<td>7</td>
<td>71.3 (5.3)</td>
<td>0.5 (0.2)</td>
</tr>
<tr>
<td>8</td>
<td>86.5 (2.3)</td>
<td>0.3 (3.9)</td>
</tr>
</tbody>
</table>

Number in parentheses is the coefficient of variation [%]

The topography of each sheet was measured using the IPST shadow Moiré system. Two examples of the topographies are given in Figure 2 below. The figure shows plots of the surface height versus position in the sheet. The plot on the left shows cockle; whereas, the plot on the right shows a wavy pattern.
The height given in the topographic plots is referenced to the best-fit plane. This means that the average value of the height data is zero. Once the topography was determined several parameters were calculated from the data. These included the maximum deviation from the height, the standard deviation of the height, the standard deviation of the gradient squared, and the volume. The average value for the three repetitions is given in Table 3.

Table 3. Summary of cockle measurements.

<table>
<thead>
<tr>
<th>Sheet</th>
<th>Maximum Height* [mils]</th>
<th>Standard Deviation [mils]</th>
<th>Standard deviation of gradient squared</th>
<th>Volume per unit area [mils]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>29</td>
<td>11</td>
<td>0.59</td>
<td>8.7</td>
</tr>
<tr>
<td>2</td>
<td>18</td>
<td>6</td>
<td>0.09</td>
<td>5.2</td>
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<tr>
<td>3</td>
<td>22</td>
<td>9</td>
<td>0.31</td>
<td>7.2</td>
</tr>
<tr>
<td>4</td>
<td>22</td>
<td>9</td>
<td>0.11</td>
<td>7.0</td>
</tr>
<tr>
<td>5</td>
<td>22</td>
<td>9</td>
<td>0.13</td>
<td>7.5</td>
</tr>
<tr>
<td>6</td>
<td>18</td>
<td>7</td>
<td>0.14</td>
<td>5.4</td>
</tr>
<tr>
<td>7</td>
<td>20</td>
<td>7</td>
<td>0.21</td>
<td>5.9</td>
</tr>
<tr>
<td>8</td>
<td>32</td>
<td>13</td>
<td>0.19</td>
<td>10.0</td>
</tr>
</tbody>
</table>

Maximum height = \((z_{max} - z_{min})/2\)
st. dev. gradient squared = \(\text{st. dev. of } [(dz/dx)^2 + (dz/dy)^2]\)
The values given in Table 3 were used to determine the sensitivity of the variables. Since we have only 6 variables but 7 possible combinations of the data, the seventh combination will be used as a measure of error. Table 4 provides a summary of the sensitivities. The numbers shown in bold indicate likely sensitivities.

Table 4. Sensitivities of Variables to Cockle

<table>
<thead>
<tr>
<th>Variable</th>
<th>Maximum Height [mils]</th>
<th>Standard Deviation [mils]</th>
<th>Standard Deviation of Gradient Squared</th>
<th>Volume per unit area [mils]</th>
</tr>
</thead>
<tbody>
<tr>
<td>CSF</td>
<td>1</td>
<td>-1</td>
<td>-0.53</td>
<td>0.3</td>
</tr>
<tr>
<td>Grammage</td>
<td>-3</td>
<td>-1</td>
<td>-0.31</td>
<td>-1.3</td>
</tr>
<tr>
<td>Forming</td>
<td>-19</td>
<td>-9</td>
<td>-0.33</td>
<td>-6.3</td>
</tr>
<tr>
<td>Pressing</td>
<td>-13</td>
<td>-9</td>
<td>+0.29</td>
<td>-6.5</td>
</tr>
<tr>
<td>Restraint</td>
<td>-23</td>
<td>-9</td>
<td>-0.83</td>
<td>-6.7</td>
</tr>
<tr>
<td>Temp.</td>
<td>19</td>
<td>7</td>
<td>0.19</td>
<td>4.9</td>
</tr>
<tr>
<td>Error</td>
<td>1</td>
<td>1</td>
<td>-0.47</td>
<td>-0.3</td>
</tr>
</tbody>
</table>

A positive sensitivity implies that the column parameter increases as the row variable goes from low to high.

Table 4 reveals that the cockle measurements were most sensitive to forming, pressing, and restraint. Sheets with modified forming, lower pressing levels, and less restraint tended to be less flat than sheets with standard forming, higher pressing levels, and more restraint. The maximum sheet height and volume per unit area were also sensitive to drying temperature. As the drying temperature increased, sheets tended to be less flat. The standard deviation of the gradient squared appeared to be sensitive to any of the variables. Freeness and grammage did not appear to be sensitive to the variables tested.

Further testing should be completed to verify these sensitivities. The effect of pressing is opposite of that expected. It was expected that a pressed sheet would
be denser, shrink more upon drying, and develop more cockle. The results indicate that pressing had the opposite effect.

Next, we determined the change in topography of the sheets due to exposure to high humidity. The sheets were exposed to 90% RH for 24 hours followed by conditioning at 20% and finally 50% RH. The topography was measured before and after the exposure to high RH. Figure 2 provides an example of before and after topographies for one of the sheets.

![Figure 2](image.png)

Figure 2. Effect of exposure to high humidity on sheet flatness.

The before and after topographies shown in Figure 2 are very similar in shape. The deformation has become more pronounced after exposure to high humidity. This behavior was also observed in a sample of machine-made copy paper that was exposed to high humidity. (Refer to last year's project report for a picture.)
Table 5 provides the percent change in both the standard deviation and the volume per unit area of the sheets. Table 5 also gives the relative sensitivities of the various parameters.

Table 5. Percent Change in Measurements Due to Exposure to High RH

<table>
<thead>
<tr>
<th>Sheet</th>
<th>% Change in St. Dev.</th>
<th>% Change in Volume per unit area</th>
<th>Variable</th>
<th>Sensitivity of % Change in St. Dev.</th>
<th>Sensitivity of % Change in Volume per unit area</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>40</td>
<td>42</td>
<td>CSF</td>
<td>32</td>
<td>25</td>
</tr>
<tr>
<td>2</td>
<td>21</td>
<td>18</td>
<td>Grammage</td>
<td>28</td>
<td>31</td>
</tr>
<tr>
<td>3</td>
<td>47</td>
<td>51</td>
<td>Forming</td>
<td>-42</td>
<td>-43</td>
</tr>
<tr>
<td>4</td>
<td>17</td>
<td>19</td>
<td>Pressing</td>
<td>-44</td>
<td>-47</td>
</tr>
<tr>
<td>5</td>
<td>35</td>
<td>33</td>
<td>Restraint</td>
<td>-94</td>
<td>-109</td>
</tr>
<tr>
<td>6</td>
<td>23</td>
<td>26</td>
<td>Temp.</td>
<td>-2</td>
<td>-1</td>
</tr>
<tr>
<td>7</td>
<td>37</td>
<td>41</td>
<td>Error</td>
<td>0</td>
<td>13</td>
</tr>
<tr>
<td>8</td>
<td>66</td>
<td>71</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The results given in Table 5 indicate that the increase in sheet cockle due to exposure to high humidity is most sensitive to restraint. Sheets that were restrained during drying experienced less change in sheet height and volume than sheets that were not restrained during drying. The modified forming process and absence of pressing both led to increased changes in sheet height and change in volume.

The results presented above indicate that restraint during drying is the most important factor in the papermaking process that affects sheet flatness. Restraint during drying produces a sheet that is flat upon drying and undergoes less change upon re-moisturization of the sheet. Beyond restraint, it appears that formation and pressing conditions will influence sheet cockle. One would expect basis weight to play a role, but the ranges in the present study may not have been large enough to reveal the effect.
We attempted to look further at the effect of restraint on drying. Sheets were dried under varying amounts of restraint by suspending the sheets in a vertical position and hanging weights from the free end of the sheet. The weights put the sheet in tension during drying. These tests did not reveal any trends when we looked at sheet height and volume. The sheet dried under no restraint appeared to have more cockle but all the sheets developed waves and wrinkles. Further work on this has not been completed.

At the Fall 1998 Paper Physics PAC meeting, it was decided that we should focus on making flat sheets that will cockle upon subsequent re-moisturization and drying. We have been developing techniques to do this, but at the present time have not been successful in getting sheets that cockle in the manner we wish. We hope to have some results to report by the Spring 1999 PAC Meeting.

We have also been developing better methods to analyze cockle. One of the capabilities that we found we needed was a method to distinguish the cockle from large waves. We have been using a Fast Fourier Transform (FFT) to analyze and eliminated the low frequency response from our data. We performed a 2-dimensional FFT on the data, eliminated the low frequency content, and calculated the inverse FFT using the modified FFT. We ended up with a filtered topography for which we can re-calculate our cockle measurements. This gives us a technique that isolates the cockle from both the curl and waves in the sheet. We are re-analyzing the data presented above on these filtered topographies at the present time. Figure 3 shows a comparison of cockle measurements before and after filtering for sheets 1 and 5 from the study. The original topography is shown in Figure 2. Sheet 1 had cockle, but sheet 2 had a wavy appearance. Figure 3 shows that the raw data did
not distinguish the two samples, but the results from the filtered data show that sheet 1 to has more severe cockle. Thus, it appears that the FFT filter can be used to eliminate the curl and waviness.

Figure 3. Effect of FFT filtering on cockle measurements.
Future Plans

We have begun a study of cockle that occurs in flat handsheets that are exposed to high humidity. We are drying the sheet under restraint and then will look at the effect of pulp freeness, formation, pressing, and drying temperature on the cockle that develops after exposure to high humidity. Before we can begin this test, we need to develop the techniques that will lead to sheets that can be dried flat and subsequently develop cockle.

After preparing this report, we feel that it is important to follow up with more experiments on the cockle that forms during drying. The results from the sensitivity study indicate that restraint, forming, pressing, and drying temperature affect the flatness of the sheet. We would like to verify and better quantify these results.
MICROMECHANICS OF FIBER NETWORKS

STATUS REPORT
FOR
PROJECT F023

Martin Ostoja-Starzewski
Jamie Castro
Michael DiMillo
Terry Bliss
Andrew N. Woods

Institute of Paper Science and Technology
500 10th Street, N. W.
Atlanta, Georgia 30318
DUES-FUNDED PROJECT SUMMARY

Project Title: MICROMECHANICS OF FIBER NETWORKS
Project Code: MICRO
Project Number: F023
PAC: Paper Physics
Division: Fiber and Paper Physics
Project Staff:
  Faculty/Senior Staff: Martin Ostoj-Starzewski (PI)
  Support: Consultant (D. Stahl)
FY 98-99 Budget: $114,312
  Allocation as Matching Funds: none
Time Allocation:
  Faculty/Senior Staff: Martin Ostoj-Starzewski (PI - 28%)
  Support: none
Supporting Research
  M.S. Students: Jaime Castro, Michael DiMillo (graduated June 1998)
  Ph.D. Students: Terry Bliss, Jaime Castro (in progress)
  External Funding: “Multiscale Mechanics of Paper,” $180,000,
                  National Science Foundation, 1997-2000.

RESEARCH LINE/ROADMAP: #11

  Improved product performance through experiments and development of quantitative
  models and algorithms.

PROJECT OBJECTIVE

  To develop an understanding of the relation of the macroscopic (scales of centimeters
  to meters) to the microscopic (fiber scales) properties of paper. Main focus on stiffness
  and strength in the MD-CD plane in terms of single fiber and fiber-fiber bond properties
  as well as formation.

PROJECT BACKGROUND

  Macroscopic and effective mechanical properties of paper in the end-use applications
  (e.g., converting operations) are a function of its microstructure. The project, therefore,
  focuses on a determination of qualitative and quantitative structure-property relations in
  paper physics and mechanics.

SUMMARY OF RESULTS (March 97--March 98)

  The ultimate goal of our research is the development of models of mechanical proper-
  ties of paper from the standpoint of a multiscale, statistical microstructure (with helical
  fibrils, fibers, flocs, etc.). The work is theoretical and experimental in nature.

IPST Confidential Information - Not for Public Disclosure
(For IPST Member Company’s Internal Use Only)
On the theoretical side, the following analyses are conducted: (i) mechanics of a single fiber treated as a layer of helically-wound fibrils; (ii) computational mechanics programs of random fiber networks which have been further developed (several versions in two or three dimensions, or with rigid versus flexible fiber-fiber bonds); (iii) modeling of the statistics and the spatial correlation structure of four elastic and strength properties measured in the conventional TAPPI tests.

On the experimental side, extensive tensile strength tests of elasticity and strength of paper have been, and are being, carried out. These tests involve specimens over a very wide range of sizes/scales: from 2mm to 2m are being carried out on conventional paper. It is found that the loading/boundary conditions, consistent with Saint-Venant’s principle, have a significant influence on effective stiffnesses and failure properties. Different effects are established for the pure size effect under geometrically similar loading conditions. Special attention is paid to the statistics of the standard TAPPI 7”x1” test versus other specimen sizes.

PROJECT GOALS FOR FY 1999-2000
1. Extension, optimization, and acceleration of the computer models of random fiber networks to handle more fibers by one or two orders of magnitude than presently possible. Systems of tens of thousands (and perhaps more) of fiber-beams are envisaged.
2. Further verification of the fiber network model using laboratory experiments.
3. Investigation of optimal formation patterns for best mechanical properties of paper.
4. Investigation of the effect of strength additives on the overall strength of paper.
5. Inclusion of single fiber mechanics for input to the computer models of random fiber networks, and further development of a model of a single fiber treating it as a filament-wound multi-layer composite.
6. Correlation of mass distribution and crack/damage patterns (a goal for the next year).
7. Biaxial tests and biaxial failure envelopes (a goal for the next two years).

Commercial Impact:
With respect to point 2 above, it is estimated that some 20% of fibers may be saved while maintaining the same strength of paper, providing optimal formation is present. Therefore, the economic benefits should be on the order of ~20%.

However, actual progress will depend on the staff and student support. The NSF-supported work of Dr. Woods is only tangentially related to this project. At present, the work of two doctoral students (Jaime Castro and Terry Bliss) is also only indirectly connected with this PAC project as it centers on the fracture mechanics and percolation of fine particles through random fiber mats, respectively.
PROJECT DELIVERABLES
1. Development of computer models for effective conductivity and mechanical (stiffness, strength, and fracture) properties of fiber networks in terms of single fiber and fiber-fiber bond properties as well as formation. This includes a unique 3-D fiber network model capable of handling thousands of interacting fibers. Indication of optimal fiber network systems.
2. Experimentally based predictions of paper stiffness, strength, and fracture (and other related variables) as functions of specimen size (over a very wide range of scales) and loading conditions. This includes a novel characterization of statistics and a first-ever characterization of correlation structure of paper webs' mechanical properties in the MD-CD plane.

PROJECT SCHEDULE

<table>
<thead>
<tr>
<th>Tasks</th>
<th>June 98</th>
<th>Sept. 98</th>
<th>Dec. 98</th>
<th>March 99</th>
<th>June 99</th>
<th>Sept. 99</th>
</tr>
</thead>
<tbody>
<tr>
<td>1: 3-D network model with flexible fiber-fiber bond deformations</td>
<td>--------</td>
<td>--------</td>
<td>--------</td>
<td>--------</td>
<td>--------</td>
<td>--------</td>
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<tr>
<td>2: 3-D network model running on workstations and supercomputers</td>
<td>--------</td>
<td>--------</td>
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<tr>
<td>3: Parametric studies of stiffness and strength using the 3-D network model</td>
<td>--------</td>
<td>--------</td>
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<tr>
<td>4: Model of a single fiber as a multilayer composite</td>
<td>--------</td>
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<tr>
<td>5: Uniaxial and biaxial tests on microsamples</td>
<td>--------</td>
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<td>--------</td>
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<tr>
<td>6: Tension strength tests with small (1&quot; and 10&quot;) and large grips (2m)</td>
<td>--------</td>
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<td>--------</td>
</tr>
</tbody>
</table>
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   4.8 Further directions

5. Significant Findings and Conclusions to Date

6. Commercial Impact

Bibliography
1. Geometry and Scale Effects in Paper
   1.1 A Hierarchy of Scales

   A starting point in our analysis is offered by the well-known multiscale structure of paper with some six principal, distinguishable levels (or scales) involved in any study of its physical/mechanical properties. With reference to Figure 1, these are:
   (a) a roll of paper on a paper machine with a possible presence of streaks—scale of meters through hundreds, thousands, ... of meters;
   (b) smallest continuum level on scales of millimeters;
   (c) a random fiber network on millimeter and smaller scales;
   (d) a single fiber with its layers P, S1, S2, and S3, and the lumen;
   (e) a fibril as a multilayer composite wound of helical fibrils;
   (f) a cellulose chain.

   Our principal focus in this micromechanics project has, in the past, centered on a passage from the spatially random fiber structure of level (c) to the continuum model of level (b)---we call it a (b)-(c) bridge. This continues to be the principal task of our PAC project (see Section 3) because, on one hand, continuum-type properties are necessary for any study of mechanics of paper as, say, a plate or a shell body on macroscopic scales (e.g., flutter, crack propagation), while formation and fiber network organization is the first level below this continuum scale.

   A second topic that was considered, especially in the last PAC report, was the (a)-(b) bridge---its investigation being based on tensile strength tests on a wide array of paper specimens, with paper coming from two very different origins. Further results, along with an ensuing analytical model are presented here as well (see Section 4).

   However, a new topic that we begin to consider here is the elasticity of a single fiber. A study of response of a ring-like bundle of helical fibrils is reported in Section 2. This begins to provide new insights into the (c)-(d) bridge.
Fig. 1 A hierarchy of scales in paper: (a) a roll of paper on a paper machine on the scale of meters with a possible presence of streaks; (b) smallest continuum level on scales of millimeters; (c) a random fiber network on millimeter and smaller scales; (d) a single fiber with its layers P, S1, S2, and S3, and the lumen; (e) a fibril; (f) a cellulose chain.
1.2 Basic geometric characteristics of fiber networks

A discussion of the planar Poisson line field (involving infinite length fibers), which is the basis of the Cox model of paper physics, has been given in the 1998 PAC report. Here we continue the line begun in the 1997 and 1998 PAC reports focusing on the finite length fiber fields, which, in the first place, allow a study of formation/flocculation effects.

Coverage: average number of fibers per point in the MD-CD plane

\[ \bar{c} = \frac{\langle \text{fibers} \rangle}{\text{point}} \] (1.1)

Fiber is taken as a rectangular-cross-section prism

\[ w \times t \times L \] (1.2)

where \( w \) is width, \( t \) is thickness, and \( L \) is length.

The test window dimensions are LMD, LCD, and LZD (x, y, and z, respectively). From these we find the fiber volume fraction

\[ v^f = \bar{c} \frac{t}{LZD} \] (1.3)

A control parameter is introduced: density \( d \) that is defined as the total fiber length (in mm) per MD-CD square millimeter area, regardless of the network thickness. By keeping \( d \) and LZD independent, we may have a paper with the same coverage but of different degrees of compaction---this would correspond to different degrees of pressing during papermaking.

We note that

\[ \bar{c} = dw \] (1.4)

The total number of all the fibers is computed by the program so as to satisfy the prescribed density and test window dimensions.

The compaction is measured by RBA defined as

\[ RBA = \frac{A_{\text{bonds}}}{A_{\text{projected}}} \] (1.5)

where \( A_{\text{bonds}} \) is the total area of all bond parallelograms, while \( A_{\text{projected}} \) is the total projected area of all fibers.

For small coverages only, there's a relationship between RBA and coverage [1]

\[ RBA = 1 - \left(1 - e^{-\bar{c}}\right)/\bar{c} \] (1.6)

When we use a small ZL-dimension for a thin network, we find that this relationship is

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quite good. However, when we use the same coverage in a thicker network, the relationship is not good any more. This makes sense, as in Deng and Dodson's work [1] they assume any two fibers which pass over the same MD-CD (i.e., x,y) point make a bond, and this is approximately true in very thin networks but not in thick networks.

Flocculation is modeled through a 'random-star' process: a number of flocs (i.e., floc centers) are generated in the given test window. This is controlled by a parameter \( n = \) flocs per unit \( (1\,mm^2) \) area in the MD-CD plane. Next, a fiber's center is generated at a distance \( r \) from the floc center according to a probability density function

\[
f(r) = \frac{b^2}{2}r + b \quad r > 0
\]

As \( b \) goes up, all fibers tend to be closely clustered into what might be called "snowflakes", while as \( b \) goes to zero, all fibers tend to scatter away. The dependence on \( b \) is shown in Fig. 2. Heavily involved in the generation of fibers is their angular density function specifying the angle \( \theta \) a fiber's axis makes with respect to the x-axis (MD). To specify this function we usually employ a Fourier series

\[
f(\theta) = \frac{1}{\pi}(1 + a_1 \cos 2\theta + a_2 \cos 4\theta + \ldots + a_n \cos 2n\theta)
\]

(1.8)

Note that other functions---such as Cauchy or von Mises---can also be employed, but (1.8) affords a number of free parameters as opposed to just one or two as in those special cases.

A fiber's random radial distance \( r \) from the parent is made proportional to the first coefficient of the angular density function \( f(\theta) \). That is, we carry out a dilation of the floc in the MD-direction \( (x) \) and no dilation in the CD-direction \( (y) \)

\[
\begin{align*}
r_x &= (1 + a_1)r \cos \theta \\
r_y &= r \sin \theta
\end{align*}
\]

(1.9)

The total number of all the fibers is divided by a random uniform mechanism between all the flocs. This is why some flocs appear to have more fibers than others, as seen in Fig. 2. One can then identify an average segment length \( \langle s \rangle \) (which is, of course, a fraction of the average fiber length \( \langle l \rangle \)) as well as an average number of bonds with other fibers per fiber. This, in fact, leads to a partition of a fiber into a chain of free fiber segments, each of which is treated as a single finite element as discussed further below.

Note that (i), on the technical side, this fiber field generation process is a special kind of a Boolean model [2], and (ii) the fibers need to be generated in a window larger by a marginal boundary layer of width \( \langle l \rangle / 2 \) so as to obtain a spatially homogeneous field.
Fig. 2(a) Effect of $b$ on generation of flocculated fields of fibers of length 1 in a square window $5 \times 5$, with 4 flocs per $1 \times 1$ area on average; $a_i = 0$ (1) in the left (right) column.
2. Elasticity of a Helical Fibril

2.1 Basic Concepts

The mechanical response of a fiber is a result of interaction of several layers (S1, S2, ...) of fibrils, each of which is characterized by a certain fibril angle or a distribution thereof. Several studies on the mechanics of a fiber treated as a cylindrical shell were conducted in the sixties and the seventies [3, 4, 5, 6, 7, 8]. However, the material of the shell was treated ab initio as an orthotropic continuum---either in two or three dimensions---rather than as a bundle of fibrils. Up to the present, several unresolved points and controversies---apparently due to the choice of boundary conditions and to a presupposition of a fiber's wall as either a 2-D or a 3-D continuum---surround those findings. In fact, even the issue of fibrils' geometry in these models is not clear.

In the first place, we observe that the fiber wall cannot contain two contiguous fibrils winding parallel on two different radii yet characterized by the same, fixed helix (or, equivalently, fibril) angle. This inconsistency may be disregarded in a thin-walled shell, but not in a thick-walled one, such as the S2 layer. As a result, a thick cell wall of fibrils having one helix angle is not simply orthotropic but must exhibit a higher level anisotropy. Next, the assumption of a constant fibril angle would contradict the assumption of a perfectly uniform (i.e., periodic) arrangement of fibrils' elliptical sections made by crossing any plane aligned with a fiber's axis. If we recall that, going through the cell wall thickness, two contiguous fibrils cannot be parallel while being characterized by the same helix angle, we conclude that the fiber wall is anisotropic rather than orthotropic.

Motivated by all these issues, we approach this problem differently: we want to derive an effective continuum-like mechanical response of a fiber from a consideration of a thin shell of fibrils. We work under the following assumptions (Fig. 3):
- the bundle is composed of \( m_2 \) fibrils, uniformly spaced along the perimeter of a circle of radius \( r_2 = R_1 + R_2 \), thus forming a ring in a plane perpendicular to the fiber axis;
- each fibril's equilibrium configuration is a helix of constant radius \( r_2 = R_1 + R_2 \) and constant helix angle \( \alpha_2 \);
- fibrils are linear elastic (with axial modulus \( E \) and Poisson's ratio \( \nu \)) and undergo very small strains only.

Now, the axial strain of a straight fiber is defined as

\[
\varepsilon = \frac{\tilde{h} - h}{h}
\]  

(2.1)

where \( h \) and \( \tilde{h} \) are the original and final lengths of the fiber, respectively (Fig. 4). The rotational strain of the fibril is defined as
\[ \beta_2 = r_2 \frac{\bar{\theta}_2 - \theta_2}{h} \]  

(2.2)

where \( \theta_2 \) and \( \bar{\theta}_2 \) are the initial and final angles that a fibril sweeps out in a plane perpendicular to the fiber's axis, respectively. Finally, the angle of twist per unit length of the fiber is defined as (note \( \beta_2 = r_2 \tau_2 \))

\[ \tau_2 = \frac{\bar{\theta}_2 - \theta_2}{h} \]  

(2.3)

Fig. 3(a) A helically shaped fibril of constant helix angle \( \alpha_2 (= 90^\circ \) - fibril angle). (b) A passage from a bundle of helical fibrils to an effective 1-D continuum.

2.2 Axial-twisting coupling and dynamic response of a single fiber

We are interested in an effective axial-torsional response of the thin shell of fibrils, that is in a relation between the axial force \( F \) and torque \( M \) on one hand, and the axial strain \( \varepsilon \) and the rotational strain \( \beta = R \tau_2 \) on the other hand, Fig. 3. Thus, if by a (generalized) force we understand the pair of \( F \) and \( M \), and by a (generalized) displacement the pair of \( \varepsilon \) and \( \beta \), this response is (e.g., [9])

\[ F = C_{11} \varepsilon + C_{12} \beta \]
\[ M = C_{21} \varepsilon + C_{22} \beta \]  

(2.4)
Here, from a requirement of a positive strain energy density, we obtain two conditions
\[ C_{12} = C_{21}, \quad C_{11}C_{22} \pm C_{12}C_{21} > 0 \] (2.5)

Evidently there are couplings between the axial and torsional responses of the fiber-shell, which are specified by four constitutive coefficients \( C_i \). In the language of continuum mechanics, the helically wound fiber is a 1-D micropolar continuum of a non-centrosymmetric (chiral composite) type (Fig. 3). On the other hand, as elaborated in our previous PAC reports, small scale, the in-plane mechanics of paper is locally described by a 2-D centrosymmetric micropolar continuum due to its fiber network structure. In 3-D, however, paper would more accurately be modeled by a non-centrosymmetric continuum: MD-CD loads cause MD-CD strains as well as anti-plane warp.

It is important to note that equations (2.4) apply to a fiber made of helically wound fibrils regardless of whether the derivation of the \( C_i \) coefficients is made from the standpoint of a theory of a fibril bundle or a continuum shell. Indeed, it was shown, in the context of structural mechanics [9], that either assumption would lead to a few percent difference (at most \(-11\%\)) for any of these coefficients. It remains to be seen what those differences would be for a shell made of thin fibrils rather than thick wires. We have recently derived the \( C_i \) coefficients explicitly, which should make such an investigation possible. In a due course, the new, extended derivation should permit an introduction of hygroscopic effects (shrinkage strains), failure of fibrils, etc.

The constitutive equations (2.4) lead us to derive a system of two coupled wave equations governing the axial-twisting response of a fiber
\[ C_{11} \frac{\partial^2 u}{\partial x^2} + C_{12} \frac{\partial^2 \varphi}{\partial x^2} = \rho \frac{\partial^2 u}{\partial t^2}, \quad C_{21} \frac{\partial^2 u}{\partial x^2} + C_{22} \frac{\partial^2 \varphi}{\partial x^2} = J \frac{\partial^2 \varphi}{\partial t^2} \] (2.6)

where \( \rho \) is the mass density and \( J \) is the mass polar moment of inertia. Now, if we consider a monochromatic wave propagation along the fiber
\[ u(x, t) = U \exp[ik(x - ct)], \quad \varphi(x, t) = \Phi \exp[ik(x - ct)] \] (2.7)

we arrive at a dispersion relation whose analysis leads to two wave speeds
\[ c_{1,2} = \frac{2(C_{11}C_{22} - C_{12}C_{21})}{(C_{11}J + C_{22}\rho) \pm [(C_{11}J - C_{22}\rho)^2 + 4\rho JC_{12}C_{21}]^{1/2}} \] (2.8)

It follows by inspection that \( c_1 < c_2 \), and, in fact, there may be an order of magnitude
difference between both wave speeds. Actual numbers depend, of course, on the choice of a theory employed for the derivation of all \( C_i \)'s or on the pertinent experiments. Given (2.7), we find that the axial vibrations of the fiber are described by two types of waves \( \text{slow and fast, each of which consists of forward and backward going pulses} \):

\[
\begin{align*}
    u(x, t) &= U_1 \exp[ik(x - c_1 t)] + U_2 \exp[ik(x + c_2 t)] + U_3 \exp[ik(x - c_3 t)] + U_4 \exp[ik(x + c_4 t)] \\
    \varphi(x, t) &= \Phi_1 \exp[ik(x - c_1 t)] + \Phi_2 \exp[ik(x + c_2 t)] + \Phi_3 \exp[ik(x - c_3 t)] + \Phi_4 \exp[ik(x + c_4 t)]
\end{align*}
\]

Next, if we consider the ratio of axial to torsional amplitudes \( U / \Phi \), we conclude (after some derivations) that the waves that are primarily axial in nature \( (U / \Phi > 1) \) propagate at speeds \( c_2 \), while the waves that are primarily torsional in nature \( (U / \Phi < 1) \) propagate at speeds \( c_1 \).

Finally, we may consider purely axial and purely torsional waves under a restrictive (and simplistic) assumption of no coupling of both motions. In that case we immediately arrive at two uncoupled wave equations

\[
\begin{align*}
    C_{11} \frac{\partial^2 u}{\partial x^2} &= \rho \frac{\partial^2 u}{\partial t^2} \\
    C_{22} \frac{\partial^2 \varphi}{\partial x^2} &= J \frac{\partial^2 \varphi}{\partial t^2}
\end{align*}
\]  

(2.10)

Again, an inspection of some typical numbers of helically wound fiber-shells suggests that the speed \( c_2 \) of primarily axial waves is on the same order as that corresponding to the speed of waves governed by \( (2.10)_1 \)---i.e., \( \sqrt{C_{11}/\rho} \)--- while the speed \( c_1 \) of primarily torsional waves is on the order of waves governed by \( (2.10)_2 \)---i.e., \( \sqrt{C_{22}/J} \).

---

Fig. 4(a) A micrograph showing the variability of fibrils’ arrangements [30].
3. Computational Mechanics of Random Fiber Networks

3.1 Three-Dimensional (3-D) Model

The geometric aspects of our modeling of mechanics of fiber networks were given in Section 1.2. The actual work proceeds through a series of models.

3.1.1 Stage 1 of the 3-D model (completion: May 1, 1998 on schedule): elasticity

There is a basic fact which casts doubt on any fiber network model in which infinitely long fibers carry axial loads only, and, therefore, fiber segments are joined by pivots, Fig. 5(a). Namely, any two cellulosic fibers have a finite contact area of hydrogen bonding [9], which would be sheared by hinge-type connections. While it is very difficult to assess experimentally to what extent this region is deformable, our basic model will treat it as somewhat deformable in the sense that bonds are rigid but have no dimension, and fiber segments are treated as extensible beams from node to node of the graph $G(V, E)$.

Here, $V$ is the set of all vertices, while $E$ is the set of all edges (fiber segments). This approach is entirely analogous to treating a frame as a system of 'welded' beams. At the second stage (Section 3.1.2) these bonds are modeled as flexible angular joints, whereby, by making the flexibility go to infinity, we recover the original, rigid-bond model.

Our modeling of mechanics of fiber networks is similar to a model of a cement-coated wood strands composite [10], and is based on the following assumptions and steps:

(i) Generate a system of finite-length straight fibers such as shown in Fig. 5(b) according to specific geometric characteristics: distribution of fiber lengths and widths, distribution of angular orientations of fiber chords, etc. The fibers are laid in three dimensions on top of one another with a possible non-zero out-of-plane angle.

(ii) Fibers are homogenous but each fiber may have different dimensions and mechanical properties, sampled from any prescribed statistical distribution.

(iii) Each fiber is a series of linear elastic 3-D extensible Timoshenko beam elements. Each of these is described by a stiffness matrix written here in an abbreviated form set up in a corotational coordinate system [11]

$$
\begin{bmatrix}
F \\
T \\
M_y^a \\
M_x^a \\
M_y^b \\
M_z^b \\
M_z^c
\end{bmatrix} =
\begin{bmatrix}
\frac{EA}{l} & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{GJ}{l} & 0 & 0 & 0 & 0 \\
0 & 0 & 4a(l^2 + 3g) & 0 & 2a(l^2 - 6g) & 0 \\
0 & 0 & 0 & 4b(l^2 + 3h) & 0 & 2b(l^2 - 6h) \\
0 & 0 & 2a(l^2 - 6g) & 0 & 4a(l^2 + 3g) & 0 \\
0 & 0 & 0 & 2b(l^2 - 6h) & 0 & 4a(l^2 + 3g)
\end{bmatrix}
\begin{bmatrix}
\Delta L \\
\Delta \theta_x \\
\theta_y^a \\
\theta_z^a \\
\theta_y^b \\
\theta_z^b \\
\theta_z^c
\end{bmatrix}
$$

(3.1)
Fig. 5. Samples of a planar Poisson line field (a) and a finite fiber field (b) generated from (1.8) with $a_1 = 1$ and all other $a_i$'s equal zero. In both cases test windows of size $L \times L$ are considered.
where
\[
\begin{align*}
g &= 12 \frac{EI_y}{GA} \quad h = 12 \frac{EI_z}{GA} \\
a &= \frac{EI_y}{l(12g + l^2)} \\
b &= \frac{EI_z}{l(12h + l^2)}
\end{align*}
\] (3.2)

Here \( F \) and \( T \) are the axial force and the twisting moment, while \( M_y^a, M_z^a, M_y^b, \) and \( M_z^b \) are the bending moments around the \( y \) and \( z \) axes at the \( a \) and \( b \) ends, respectively (Fig. 6(c)). Also, \( \Delta L, \Delta \theta_x, \theta_x^a, \theta_z^a, \theta_y^b, \) and \( \theta_y^b \) denote the corresponding kinematic quantities: axial elongation, angle of twist, and four angles of rotation. Finally, \( l, A, J, I_x, \) and \( I_y \) are, respectively, the length, cross-sectional area, cross-sectional polar moment of inertia, and the moments of inertia with respect to the \( x \) and \( y \) axes. \( E \) and \( G \) are the Young's modulus and shear modulus of a fiber-beam.

Fig. 6 (a-b) A 3-D fiber network (after [12]) and its model dilated in ZD. (c) A 3-D Timoshenko beam element modeling a single fiber segment between two consecutive bonds with other fibers.

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(iv) All the intersection points are identified so as to set up a connectivity matrix.
(v) Solve for the equilibrium subject to all the constraints and kinematic (essential) boundary conditions ($\bar{\varepsilon}_{ij}$ is the volume average strain)

$$u_i = \bar{\varepsilon}_{ij}x_j$$

(vi) All six effective, in-plane stiffness coefficients are determined from the postulate of equivalence of strain energy stored in a square-shaped window of finite thickness with the strain energy of an equivalent, hypothetical continuum.
(vii) Note that the traction boundary condition

$$t_i = \bar{\sigma}_{ji}n_j$$

cannot be applied in an unambiguous way to a beam network, so as to infer the effective compliances directly from a postulate of complementary energy equivalence. Such a problem has earlier been discussed in the setting of disordered central force networks [13].

The current limitations of this model are as follows:
- maximum number of nodes: 7344
- maximum number of fiber segments: 11016
- maximum number of degrees of freedom: 44064
- maximum number of fibers: 1614
- maximum number of flocs: 500

For the sake of completeness of presentation, we now repeat here the simulation results from our PAC Report 1998 that point to the importance of rigidity of fiber-fiber bonds. The undeformed network, shown in Fig. 5(b) in its top view, has the following parameters: window size: 4x4x0.1 mm $a_1 = 1$, and other coefficients in equation (1.1) are zero; fiber length: 2 mm; fiber width: 0.04 mm; fiber height: 0.015 mm. As a result of a Monte Carlo fiber placement process, we obtain: 195 fibers with an average of 4.8 bonds per fiber, the whole system having 859 nodes with six degrees of freedom per node.

**Example #1**

A state of deformation corresponding to axial strain $\varepsilon_{11} = 8\%$ is shown in Fig. 7(a). The analyzed strain is actually 1%, and displacements are magnified for clarity. Compare this deformed network to that in Fig. 7(b), which shows the same network of fibers subjected to the same strain but with the ratio of fiber flexural stiffness to fiber axial stiffness reduced by a factor of $10^{-4}$. Note the following:
Fig. 7(a) Deformation of the network of Fig. 5(b), with 195 originally straight fibers, and fiber bending (and shear) present, under axial strain $\epsilon_{11} = 1\%$; (b) the same network, with fiber bending almost absent, subjected to same strain; all displacements magnified by a factor 8 for clarity. Figure (b) shows large, mechanism-type motions of the network including those of some fibers which spring outside the original domain of the network.
(i) The sharp kinks we see in both figures are primarily the artifact of simple computer graphics---the micromechanical model assumes fibers deform into almost continuous curves with slight kinks caused by shear alone. Magnification creates the appearance of large displacements---actually an infinitesimal displacement assumption is used in the derivation of the fiber stiffness.

(ii) The kinks are far more pronounced when fibers have low flexural stiffness. Portions of the network where connected fibers do not form triangular pores can generate significant forces in response to deformation when fibers have high flexural stiffness, but they cannot do so when fibers rely almost entirely on axial stiffness. These portions of the network are not stable in the sense of loss of generic rigidity that is discussed in Section 3.3 below.

(iii) The experimentally observed fiber network deformations reported in the PAC 1998 Report are qualitatively similar to those of Fig. 7(a).

Example #2
This example focuses on the effect of formation (flocculation) on the change of stiffness. The formation is studied by varying the flocculation parameter $b$ of the random star model of Section 1.2. Thus, in Fig. 8 two networks are shown: (a) with $b = 0.2$ and (b) with $b = 1.6$. For clarity, only a thin layer is analyzed, and only fiber axes (no fiber widths) are shown. The networks have $RBA = 0.288$ and $RBA = 0.327$, respectively. More information on the example is provided below.

<table>
<thead>
<tr>
<th>Case (a)</th>
<th>Case (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Test area LMD $\times$ LCD $\times$ LZD:</td>
<td>5mm $\times$ 5mm $\times$ 0.1mm</td>
</tr>
<tr>
<td>Fiber density</td>
<td>40</td>
</tr>
<tr>
<td>Flocs per 1 mm$^2$ of MD-CD area:</td>
<td>1</td>
</tr>
<tr>
<td>Fiber length:</td>
<td>1mm</td>
</tr>
<tr>
<td>Fiber width:</td>
<td>0.05mm</td>
</tr>
<tr>
<td>Fiber thickness:</td>
<td>0.015mm</td>
</tr>
<tr>
<td>Total number of fibers:</td>
<td>1,200</td>
</tr>
<tr>
<td>Fiber angular distribution:</td>
<td>$f(\theta) = (1 + \cos 2\theta)/\pi$</td>
</tr>
<tr>
<td>Average number of bonds per fiber:</td>
<td>5.1</td>
</tr>
<tr>
<td>RBA:</td>
<td>0.288</td>
</tr>
<tr>
<td>Fiber Young's modulus $E^f$:</td>
<td>50,000 MPa</td>
</tr>
<tr>
<td>Fiber Poisson's ratio $\nu^f$:</td>
<td>0.3</td>
</tr>
<tr>
<td>Fiber strength:</td>
<td>100 MPa</td>
</tr>
</tbody>
</table>

In both cases, the basis weight = 22g/m$^2$ and coverage = 2 (because there is the
same number of fibers), but the RBA strongly depends on the sheet thickness: with \( LZD \) decreasing, \( RBA \) is increasing. Next, we note that as the flocculation increases: for MD: stiffness decreases by \(-15\%\), strength increases by \(-10\%\), for CD: stiffness decreases by \(-9\%\), strength decreases by \(-40\%\).

(a) 

![Network Example #2 with \( b = 0.2 \)](image)

(b) 

![Network Example #2 with \( b = 1.6 \)](image)

Fig. 8. A network of Example #2 with (a) \( b = 0.2 \) and (b) \( b = 1.6 \).
3.1.2 Stage 2 of the 3-D model (completion: February 1, 1998): springy fiber-fiber bonds and progressive damage

The work focused on a modification of the progressive failure algorithm to account for a relative fiber-fiber bond deformation by introducing angular, linear elastic springs at the bond between two interacting fibers. As a result, a point at the fiber-fiber intersection in the MD-CD plane really represents two points---one belonging to the fiber on top, and the other one to the fiber on the bottom (Fig. 9). Let us consider an undeformed configuration of two fibers, taken for simplicity to be straight and crossing at an angle $\alpha_0 = 90^0$. We next consider two possibilities: one when $\alpha_0$ is preserved, and another when $\alpha_0$ becomes $\alpha$. The first case corresponds to a frame model of structural mechanics.

Fig. 9(a) Two, initially straight fibers, crossing at $\alpha_0 = 90^0$. (b) Deformation without the angle change. (c) Deformation with the angle change.

A major advantage of having this angular spring in the model, is the possibility of debonding of both fibers. This may be carried out for flexible angular springs, as well as for very rigid ones. Thus, effectively, we add a brittle bond failure mechanism due to shear stresses in addition to that of fiber segment failure.
Example #3

test area: 2mm x 2mm x 0.08mm
density 25.6
flocs per 1 mm$^2$ of MD-CD area: 1
fiber length: 1mm
density 25.6
fiber width: 0.04mm
fiber thickness: 0.015mm
total number of fibers: 170
fiber angular distribution: $f(\theta) = \frac{1}{\pi}$ (i.e., isotropic)
average number of bonds per fiber: 3.26
fiber Young's modulus: 50,000 MPa
fiber Poisson's ratio: 0.3
fiber bond modulus: 10,000 MPa
fiber strength: 100 MPa
failure by fiber fracture only, no fiber-fiber debonding in both cases

The elastic properties of the equivalent continuum, $E_x$, $E_y$, $G_{xy}$, etc., can be extracted from the 6x6 continuum compliance matrix. Here elastic modulus in MD = 42.8 (units discussed in next paragraph) and $v_{xy} = 0.338$.

Note:
(i) Our use of the terms stress and strain as they affect the definition of the terms in the compliance matrix. When referring to the volume occupied by the network, we typically calculate strain as an overall displacement divided by an original length. Stress is the sum of nodal forces on a face in a given direction divided by the length of the face, not divided by the thickness of the volume.
(ii) The final value of the elastic modulus may be significantly less than that extracted from the virgin network's response.

Figure 10 shows the network after several displacement increments. Elements that have failed are shown in bold. Note that fiber ends on the bottom and top faces of the test win-
dow are free to translate or rotate in any direction.

Fig. 10. The deformed network with failed fiber elements shown as heavy lines.

The stress-strain history is shown in Fig. 11. For the completeness of presentation we also give a sequence of stresses and strains corresponding to twelve consecutive loading increments:

<table>
<thead>
<tr>
<th>INC</th>
<th>STRAIN</th>
<th>STRESS</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>.238710E-03</td>
<td>.548900E-02</td>
</tr>
<tr>
<td>2</td>
<td>.238710E-03</td>
<td>.400926E-02</td>
</tr>
<tr>
<td>3</td>
<td>.318493E-03</td>
<td>.534926E-02</td>
</tr>
<tr>
<td>4</td>
<td>.318493E-03</td>
<td>.477932E-02</td>
</tr>
<tr>
<td>5</td>
<td>.330021E-03</td>
<td>.495231E-02</td>
</tr>
<tr>
<td>6</td>
<td>.330021E-03</td>
<td>.396624E-02</td>
</tr>
<tr>
<td>7</td>
<td>.475324E-03</td>
<td>.571250E-02</td>
</tr>
<tr>
<td>8</td>
<td>.475324E-03</td>
<td>.549228E-02</td>
</tr>
<tr>
<td>9</td>
<td>.524694E-03</td>
<td>.606275E-02</td>
</tr>
<tr>
<td>10</td>
<td>.524694E-03</td>
<td>.573700E-02</td>
</tr>
<tr>
<td>11</td>
<td>.609455E-03</td>
<td>.666377E-02</td>
</tr>
<tr>
<td>12</td>
<td>.609455E-03</td>
<td>.642758E-02</td>
</tr>
</tbody>
</table>

Dots at the tops of vertical segments are microfailures, and dots at the bottom of vertical segments are not. Note that the tangent stiffness decreases with each microfailure, and that the tangent stiffness is always equal to the secant from the origin---even with damage, the network is linear elastic and will return to zero strain if the stress is lowered to zero. Notice that immediately after a failure, the strain is not increased. This creates the vertical drops in the stress-strain plot.
Fig. 11. Stress-strain history of the network.

- On the relations between fiber and bond properties and network properties:
  (i) One network was used to investigate the effects of bond and fiber stiffness and strength properties on network behavior. The importance of bond stiffness increases when fibers are not continuous across the network because the fibers cannot offer a load path independent of bonds. A network size of 2\text{mm} \times 2\text{mm} square with a fiber length of 1\text{mm} was used. The alignment constants were all equal to zero. The bond and fiber area properties were set to be constant.
  (ii) The network was analyzed under several sets of conditions to determine the relative effects of fiber and bond stiffness on network stiffness. Fiber modulus of elasticity $E$ was held constant at 50,000 \text{MPa}, and bond $E$ was varied from 10 \text{MPa} to 100,000 \text{MPa}. Note that because bond geometric properties are based on a somewhat arbitrary idealized shape, we should not expect bond $E$ to correspond to values in the literature. Figure 3 shows that if bond $E$ is above 1000 \text{MPa}, network stiffness is the same as if bonds are rigid. With bond $E$ at 100 \text{MPa}, network stiffness is about two-thirds of what it is with rigid bonds. The data point labeled “Rigid bonds” is the result of the basic version of the model (Section 3.1.1) in which bonds are not flexible.
Fig. 12. Effect of bond stiffness on network stiffness (fiber $E = 50,000$ MPa).

The same network was analyzed with different conditions to evaluate the effect of bond and fiber properties on network stress-strain history. Keep in mind that because bond geometric properties are based on a somewhat arbitrary idealized shape, we should not expect bond strength to correspond to values in the literature. The following input properties were used:

**Table 1.** Input for trials (Percent Coefficient of Variation for fiber properties in brackets)

<table>
<thead>
<tr>
<th>No.</th>
<th>Fiber $E$ (MPa)</th>
<th>Bond $E$ (MPa)</th>
<th>Fiber strength (MPa)</th>
<th>Bond Strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>50000 [0]</td>
<td>100</td>
<td>100</td>
<td>0.4 [0]</td>
</tr>
<tr>
<td>2</td>
<td>50000 [0]</td>
<td>1000</td>
<td>100</td>
<td>1.2 [0]</td>
</tr>
<tr>
<td>3</td>
<td>50000 [0]</td>
<td>1000</td>
<td>100</td>
<td>2.4 [0]</td>
</tr>
<tr>
<td>4</td>
<td>50000 [0]</td>
<td>100</td>
<td>100</td>
<td>1.2 [0]</td>
</tr>
<tr>
<td>5</td>
<td>50000 [0]</td>
<td>1000</td>
<td>100</td>
<td>1.2 [0]</td>
</tr>
<tr>
<td>6</td>
<td>50000 [0]</td>
<td>1000</td>
<td>100</td>
<td>1.2 [0]</td>
</tr>
<tr>
<td>7</td>
<td>50000 [0]</td>
<td>100</td>
<td>100</td>
<td>1.2 [40]</td>
</tr>
</tbody>
</table>

Figure 13 shows the network stress-strain plots (recall the definition of stress, which was given earlier) for trials 1 through 6. Each trace has a few symbols on it to help differentiate the six. First note that the initial slopes of trials 1, 3, and 5 (dotted lines) are identical, as are those of 2, 4, and 6 (solid lines). Note also that increased stiffness of the latter group is due to the higher value of bond $E$. All of the microfailures in trials 1 and 2 are...
bond failures; all in trials 5 and 6 are fiber failures; trials 3 and 4 included a mix of bond and fiber failures. Two observations are made:

1. A ratio of fiber strength to bond strength of approximately 80 to 1 results in a mix of bond and fiber microfailures. This ratio is presumably dependent on the fiber and bond area properties used in the trials, and is not a general rule.

2. More flexible bonds appear to create a slightly more ductile response---with more network stiffness and strength preserved after the first microfailures create a drop in stiffness.

Fig. 13. Network stress-strain plots for trials 1 through 6.

A notable aspect of all six plots in Fig. 13 is the lack of significant nonlinearity prior to the peak stress. In trials 5 and 6 the first microfailure represents the peak stress---so there is no nonlinearity at all. Trials 1 and 3 seem to have the most interesting plots, with the most nonlinearity prior to the peak (trial 1) or the most ductility (trial 3) after the peak. Previous experience indicates that nonlinearity is increased by increasing the variability of fiber strength. Trial 7 is a repeat of trial 3 but with fiber strength given a Gaussian distribution with coefficient of variation equal to 40%. The stress-strain plots for trials 3 and 7 are shown in Figure 14. Note that increasing the nonlinearity by increasing fiber strength variability has led to a decrease in the ductility.
In conclusion, adjustments to fiber and bond properties lead to obvious changes in network stiffness, strength, and stress-strain histories. The user can modify the input parameters in various ways. Here lies the great power and problem with micromechanical models: The developer can try to mimic the important mechanical behavior of the real microstructure, but as more detail is added to the model, physical and mechanical properties of the constituents are known with less certainty. One example of this in our program is the bond elasticity: if we want to make the bonds flexible, we create an idealization of bond flexibility. However, we have no experimental data to provide the input so we make it up arbitrarily; we adjust the bond elasticity until overall network properties match paper properties. We must recognize that this is now simply an empirical adjustment, as opposed to a rationally determined input parameter.

![Network stress-strain plots for trials 3 and 7.](image)

**Fig. 14.** Network stress-strain plots for trials 3 and 7.

3.1.3 Stage 3 of the 3-D model (completion: January, 1999): program acceleration and optimization

The 3-D computer model was adapted to run on our Silicon Graphics computer. This task is continuing so as to lead to an order of magnitude improvement over the present limits (listed in Section 3.1.1) by a factor of 5 to 10. Another option which is currently being investigated is the possibility of running it on a supercomputer.
3.2 A Simplified Two-Dimensional (2-D) Model (estimated completion: May, 1999)

In its fullest version, our program places heavy demands on the computer memory---six degrees of freedom per each fiber segment's end, so that, in the fullest version of flexible bonds, there are twelve degrees of freedom per each fiber-fiber bond. This, in turn, leads to a necessity of solution of very large algebraic problems. Thus, we are led to a question: can a 3-D model, restricted to 2-D motions, do a satisfactory job in approximating what is predicted by a full 3-D model outlined above?

By a restriction to 2-D motions we mean a fiber network in which each and every fiber segment end is restricted to three degrees of freedom: two displacements (in MD and CD directions) and an in-plane rotation. As a result, we would have six degrees of freedom per each fiber-fiber bond in the flexible fiber-fiber bond model (Section 3.1.2).

Next, we consider a situation of rigid bonds, in which case, each and every fiber-fiber bond would be rigid (Section 3.1.1)---i.e., a total of three only---this then would lead to a number of degrees of freedom smaller by a factor two.

This entire issue of approximating 3-D problems by the 2-D ones of the same fiber network topologies is presently under study.

3.3 A comparison of the computational mechanics model to the Cox model

Our model admits the displacements and rotations of fiber-fiber bonds as possible degrees of freedom. Thus, the strain energy stored in the network is a combination of the energies due to extension/compression, shear, bending, torsion of fiber segments, as well as bond-angular (in case of flexible rather than rigid fiber-fiber bonds) interactions

\[ U = U_{\text{central}} + U_{\text{shear}} + U_{\text{moment}} + U_{\text{torsion}} + U_{\text{bond}} \]  

(3.5)

The classical Cox model admits only the energy due to axial interactions (i.e., along the fibers), and so, the strain energy is

\[ U = U_{\text{central}} \]  

(3.6)

However, \( U_{\text{central}} \) in (3.5) is different (i.e., smaller) from \( U_{\text{central}} \) in (3.6), since the former is computed from the equilibrium equations of the entire system of interacting fibers, while the latter is obtained from a postulated uniform strain field. Thus, the total energy \( U \) in (3.6) is overestimated in that \( U_{\text{central}} \) is too high, but, simultaneously, it is underestimated in that \( U_{\text{central}} \) only is present.

A quantitative comparison of all the energies is possible, but unwieldy. This is due to a need to introduce special do-loops to our program so as to compute \( U_{\text{central}} \), \( U_{\text{shear}} \), \( U_{\text{moment}} \), \( U_{\text{torsion}} \), and \( U_{\text{bond}} \) separately. Furthermore, we would have to write an addi-
tional program to solve the mechanics of a network of fibers carrying axial loads only. As explained in Section 3.4 below, the latter network would actually behave as a mechanism, which would lead to unstable behaviors making an elastostatic analysis difficult.

We can, however, readily carry out a comparison of the effective elastic moduli of both approaches. Thus, recalling (1.3), the Young’s modulus resulting from the Cox model is expressed as

\[ E^{Cox} = \frac{1}{3} V_f E^f = \frac{1}{3} Z \frac{t}{LZD} E^f \]  \hspace{1cm} (3.7)

while that resulting from the network theory is \( E^{network} \). A full comparison of all the parameter dependencies is underway and will be reported later. The basic trends are:

(i) as the \( t/LZD \) ratio goes down (i.e., sheet compaction increases), \( E^{network}/E^{Cox} \) tends monotonically towards 0.5 from below;

(ii) an increase of the fiber length at a fixed window size brings \( E^{network}/E^{Cox} \) further up;

(iii) an increase in flocculation \( b \) going up) lowers \( E^{network}/E^{Cox} \).

Example #4

Here we compare the kinematics of a network undergoing true displacements with the kinematics of a network (of identical geometry and connectivity) that corresponds to the uniform strain. An undeformed network (Fig. 15(a)), of a test area 3mm x 3mm and density \( d = 40 \), is subjected on its boundaries to an \( \varepsilon_{xx} \) strain, Fig. 15(b). The displacements are calculated here by computational mechanics; recall Section 3.1.1. The comparison with a uniform strain model is carried out in Fig. 16. The first of these---i.e., 16(a)---gives the path each node takes from its original position to the displaced position of Fig. 15(b). Note that any deviation from a horizontal line, or any length that is not proportional to a node’s original \( x \) position, represents the effect of non-uniform strain.

Figure 16(b) shows the difference between each node’s displacement in the present analysis and what its displacement would be if a uniform strain---equation (3.3)---were imposed not only on the exterior, but also on its interior nodes. The small dot is the displaced position of each node under a uniform strain throughout the network. If there does not appear to be a line attached to the dot, then the two displacements are approximately the same. The longer the line attached to the cross, the more significant the difference between the two displacements. Note that the circular shape slightly above and to the left of the middle is a vortex, with nodes on the top of the vortex displacing less, and the nodes on the bottom displacing more than the uniform strain assumption would suggest.
Fig. 15(a). An undeformed fiber network, and (b) its deformation under an $\varepsilon_{xx}$ strain.
Fig. 16(a) A display of the path each node of Fig. 15(a) takes from its original position to the displaced position of Fig. 15(b). (b) A display of the difference between each node's displacement in the present analysis and what its displacement would be if a uniform strain were imposed not only on the exterior, but also on its interior nodes.
3.4 The Limiting Case of Infinitely Long, Axial Load Carrying Fibers

Inspecting Fig. 7(a), we may identify the fiber segments to be edges of an edge set \( E \), and pivots to be vertices of a vertex set \( V \). The fiber network is therefore represented by a graph \( G(V, E) \). A necessary condition for any planar graph to be isostatic (i.e., the minimum condition for it not to be a mechanism) is provided by the structural topology (e.g., [14]): the minimal number of edges should be

\[
|E| = 2|V| - 3
\]  
(3.8)

where \(| \cdot |\) denotes the number of elements in a given set. In fact, there are further conditions (sufficient ones) to ensure that a graph is isostatic. Edges in an isostatic graph work independently to produce rigidity; the structure is not overbraced. The removal of any single edge introduces some motion (i.e., renders it a mechanism). An example of a graph that is isostatic is given in Fig. 16.

The Cox model starts out with a network of lines spanning the entire window. Its geometry is, therefore, rigorously given by a so-called Poisson line field, e.g., [2]. It is known that, whether the lines' angles \( \theta \) are sampled from an isotropic or an anisotropic probability density function, the triple-fiber intersections occur with probability zero. Thus, we typically have vertices of connectivity 4 (i.e., with four fiber segments incident onto a vertex)---they belong to a set \( V_4 \).

![Fig. 17. A graph that is isostatic.](image)

Now, with reference to Fig. 7(a), which shows a typical realization of the Poisson line field, we see that there are two types of edges: those in direct contact with the square shaped window on which the kinematic boundary condition (3.3) is applied, and those in the interior. Clearly, the square window is needed to prevent these boundary layer edges...
from dangling---this immediately renders the entire network a mechanism. However, one may argue that the boundary layer of dangling edges is very thin, and examine condition (2.1) with respect to the graph $G(V, E)$ representing the interior network of edges not directly in contact with the square window boundary; these are shown in bold in Fig. 1(a).

Here we observe that, while vertices of $V_4$ occur in the interior of this graph, its boundary involves 2- and 3-connected vertices of sets $V_2$ and $V_3$. Now, since there are two vertices to every edge, we calculate the total number of edges in the bold drawn graph $G(V, E)$ according to

$$|E| = |V_2| + \frac{3}{2}|V_3| + 2|V_4|$$

(3.9)

Evidently, since $V = V_2 \cup V_3 \cup V_4$, the total number of all the vertices is

$$|V| = |V_2| + |V_3| + |V_4|$$

(3.10)

so that $|E| < 2|V| - 3$ and (3.5) is not satisfied---the system is underconstrained (i.e., not isostatic): it is a mechanism. Given this observation, the Cox model is not a valid model of paper or any other solid material. It is a material below a so-called rigidity percolation, a concept encountered in condensed matter physics [15].

Now, the Cox model does give finite values for elastic moduli including the shear modulus because the fibers span the entire test window. The situation is analogous to a square lattice truss, which, even though it is an obvious mechanism (as it violates (3.5)), will give finite orthotropic moduli if subjected to (3.3). However, both systems respond as mechanisms when subjected to shear traction boundary conditions (3.4). Yet, the ability to support (quasi-conservative) shear stresses is a common definition of a solid material [18].

In real networks, fibers have finite length, so their ends are loose. When fiber ends are removed to eliminate the obvious mechanism, the number of vertices in sets $V_2$ and $V_3$ increases. Consequently, relation (3.8) is even further away from being satisfied.

Paper exhibits finite stiffnesses in 2-D as well as in 3-D. When working in 3-D, condition (3.5) is replaced by an even more stringent one as one needs more constraints when dealing with the additional degrees of freedom [14]. The paradoxical behaviors plaguing the Cox model are immediately eliminated if one replaces pivots with rigid or springy (flexible) bonds, and allows fiber segments to carry shears and bending moments in addition to axial forces---recall Section 3.1. The extension of this to three dimensions requires that fibers also carry torsional moments---recall equation (3.1).
3.5 Conclusions
3.5.1 A classification of fiber network models

There are, in general, three types of paper mechanics models. This discussion is written not as a criticism of work by others, but as a recognition of the tremendous challenges in paper physics that we are grappling with.

(i) Effective medium models based on the uniform strain assumption

These models date back to Cox [16]; see also [17]. They involve kinematically admissible but statically inadmissible fields and, thus, cannot include such salient features of paper microstructure as nonuniform stress/strain distributions due to floculation. The original Cox model assumes fibers in the paper to be infinitely long (Poisson line field geometry, Fig. 7(a) while lacking any mutual interaction, and requires an external rigid frame so as to result in an overall stiffness. This may be justifiable for a textile, but, by definition, a solid material does not require a rigid frame to support external loads. Indeed, microscale interactions between constituents (atoms, molecules, crystals, etc.) of a solid are the essence of its mechanics. Furthermore, a solid material is distinguished from a fluid one by its ability to support shear stresses.

(ii) Phenomenological deterministic and probabilistic models

The deterministic models are exemplified by the work of Lu et al [19-20]---they are typically based on the concept of a unit cell in a paper material idealized as a periodic composite; see also [22]. Clearly the mechanics is correct, but the geometric simplification---and hence, the connection to the real microstructure---is untenable. Models of the probabilistic type [23, 24] represent an opposite modeling strategy: they work with a random (i.e., non-periodic) fiber microstructure but ignore fiber-fiber interactions and oftentimes depend on the assumption of perfectly straight running cracks.

(iii) Computational mechanics models

Models of this category [25, 26] simulate the network mechanics with all interactions directly: they are consistent with the logic of planar/spatial kinematics, and can account for a multitude of fiber properties, bond properties, and geometric arrangements including floculation. These benefits come at the price of solution by extensive computer simulations, rather than closed form equations. However, with the dramatically increasing availability of computing power they allow one (even on a portable computer) to assess quantitatively the elastic and fracture properties of fiber networks, including scaling and statistics thereof.
4. Experimental Tests of Elasticity and Strength

4.1 Experimental tension strength tests for a wide range of sizes from 2mm to 2m

It is well known that the basic TAPPI test is carried out on a 7"x1" specimen. There have been some investigations on the effects of size on strength of paper [39], or the statistics of strength [40]. However, no single study appears to have covered these two aspects from a unified perspective, and, apparently, no one has looked at the local (nor global) correlation structure of several basic properties measured in the strength tests. The latter issue is analogous to the question of the correlation structure in fluid dynamics, and is highly relevant in the case of paper, given its formation on the wire. In fact, paper structure can, to some extent, be regarded as a “frozen-in turbulence.”

Let us consider the following questions:

Q1: What can the 7"x1" TAPPI test say about the web strength on a paper machine?
This can be rephrased as:
Q2: How do the paper properties depend on the specimen size and loading conditions?
Next, considering the unavoidable uncertainty, we should ask:
Q3: What is the statistical scatter and the cross-correlations of basic mechanical properties listed below (e.g., Fig. 18)?

- \( E \), elastic (Young's) modulus in lbf/in
- \( \sigma_{max} \), tensile breaking strength in lbf/in
- \( \varepsilon_{max} \), strain to failure in %
- \( TEA \), tensile energy absorption in lbf/in

and, given the turbulence origin of paper:
Q4: What is the spatial correlation structure of these four properties?

Fig. 18. Results of ten Instron strength tests showing scatter in \( E \), \( \varepsilon_{max} \), \( \sigma_{max} \), and \( TEA \).
In principle, we should ask all these questions with respect to the MD and CD properties.

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In light of Q1, we currently focus on the MD performance. Figure 19(a) presents a schematic of an array of specimens, cut in such a way that the records were kept of the relative positions of all the specimens. This was done so as to assess the correlation structure in the MD-CD plane. Next, Figure 19(b) shows the entire range of specimen sizes subjected to the tests, all in MD. For completeness, these sizes are collected in Tables 2 and 3, respectively, for paper from the Repap Technologies, Inc. and Champion International Corp. Here \( w \) stands for width and \( l \) for length (in inches). The numbers in each case we give (number of specimens in MD) x (number of specimens in CD).

### Table 2: Repap Technologies, Inc.

<table>
<thead>
<tr>
<th>( w )</th>
<th>( l = 1 )</th>
<th>( l = 7 )</th>
<th>( l = 28 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( w = 1/8 )</td>
<td>2x100</td>
<td>2x100</td>
<td>5x20</td>
</tr>
<tr>
<td>( w = 1 )</td>
<td>25x8</td>
<td>10x20</td>
<td>20x5</td>
</tr>
<tr>
<td>( w = 4 )</td>
<td></td>
<td>25x4</td>
<td>50x2</td>
</tr>
<tr>
<td>( w = 10 )</td>
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<tr>
<td>( w = 40 )</td>
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<td></td>
<td></td>
</tr>
</tbody>
</table>

### Table 3: Champion International Corp.

<table>
<thead>
<tr>
<th>( w )</th>
<th>( l = 1 )</th>
<th>( l = 7 )</th>
<th>( l = 28 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( w = 1/8 )</td>
<td>2x100</td>
<td>2x100</td>
<td>4x25</td>
</tr>
<tr>
<td>( w = 1 )</td>
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<td>8x25</td>
<td>10x10</td>
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<td></td>
<td>25x4</td>
<td>25x4</td>
</tr>
<tr>
<td>( w = 10 )</td>
<td></td>
<td></td>
<td>30x1</td>
</tr>
<tr>
<td>( w = 79 )</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Prior to the tests, all the specimens were stabilized in a standard humidity environment, and then the tests were conducted at crosshead speeds given in Table 4.

### Table 4:

<table>
<thead>
<tr>
<th>specimen size</th>
<th>1x1/8</th>
<th>1x1</th>
<th>7x1/8</th>
<th>7x1</th>
<th>7x10</th>
<th>28x1</th>
<th>28x4</th>
<th>28x10</th>
<th>28x40</th>
<th>28x79</th>
</tr>
</thead>
<tbody>
<tr>
<td>crosshead speed</td>
<td>0.142</td>
<td>0.142</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>4.0</td>
<td>4.0</td>
<td>4.0</td>
<td>4.0</td>
<td>4.0</td>
</tr>
</tbody>
</table>
Fig. 19 Schematic of an array of specimens (a) and various sizes (b) tested for strength.
4.2 Random field of mechanical properties

The degree of variability in the stress-strain response of paper is displayed in Fig. 18, which shows typical results of ten tensile tests. All the specimens are cut out of a large (several meters wide and many kilometers long) paper web. Thus, at every point, given a chosen size and shape of a test specimen, the four conventionally measured parameters (elastic modulus $E$, breaking strength $\sigma_{\text{max}}$, strain to failure $\varepsilon_{\text{max}}$, and tensile energy absorption $\text{TEA}$) are random variables. Clearly, these variables are functions of position, and so, they constitute a four-component vector random field $\mathbf{u}$

$$\begin{bmatrix} u_1 \\ u_2 \\ u_3 \\ u_4 \end{bmatrix} \equiv \begin{bmatrix} E \\ \sigma_{\text{max}} \\ \varepsilon_{\text{max}} \\ \text{TEA} \end{bmatrix} : B \times \Omega \rightarrow \mathbb{R}^4 \quad B \subset \mathbb{R}^2 \quad (4.1)$$

Here $B$ is the body domain of the paper web in the $x, y$-plane, while $\Omega$ is the probability space (or space of elementary events $\omega$).

The physical meaning of the space $\Omega$ is clarified by considering any single event $\omega$. The occurrence of this $\omega$—and hence, of specific values of field $\mathbf{u}$ that correspond to it—represents an uncertain phenomenon, which can only be described statistically. Our problem formulation is analogous to the one encountered in turbulence theories ([32, 33]), where one cannot predict exactly (i.e., deterministically) the velocity flow field and has to resort to probability tools.

Indeed, our random field of paper properties has its origin in the turbulent settling of disordered fiber suspensions on the wire, and is interpreted as a final result of a “frozen-in turbulence.” The field $\mathbf{u}$ is parametrized by the size $L_x \times L_y$ of specimens—a concept again analogous to the turbulence theory, where the sampling of values of the velocity field is conducted over certain finite volumes corresponding to the resolution of a given instrument, e.g., radar, or laser Doppler. Here, $x$ and $y$ correspond to the machine and cross directions, respectively. In the following, we focus on the $7'' \times 1''$ specimen sizes, which represent the most common paper industry standard [35]. The unnotched specimens are subjected to quasi-static tensile tests in the machine ($x$) direction. They are cut from a $79''$ (2 m) wide roll of paper provided by Champion Intl. Corp. The paper has basis weight of $\sim 21 \text{ lb/1000 ft}^2 (\sim 34 \text{ g/m}^2)$ and a caliper of $\sim 2.6 \text{ mils} (\sim 6.6 \cdot 10^{-5} \text{ m})$; the sheet contains some mechanical pulp.

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Fig. 20. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for an $8 \times 25$ array of 7” x 1” specimens. The ranges and assignments of values are shown in the respective insets.
On the theoretical side, the random field \( \hat{u} \) is, most fundamentally, specified by a family of all the finite-dimensional \( m \)-point distribution functions \( (x \equiv (x, y)) \)

\[
P\{ U_1(x_1) \leq u_{11}, \ldots, U_1(x_m) \leq u_{1m}, \ldots, U_4(x_1) \leq u_{41}, \ldots, U_4(x_m) \leq u_{4m} \} \quad (4.2)
\]

Now, in view of the tremendous amount of information that would need to be collected to obtain (4.3), we content ourselves with an assessment of the one-point statistics and of the second-order correlation structure. These are obtained through tensile strength tests on an \( 8 \times 25 \) array of our \( 7" \times 1" \) specimens. Figures 20(a)-(d) display the results of these tests for \( E, \sigma_{\max}, \varepsilon_{\max} \), and \( TEA \), respectively.

### 4.3 Statistics and Correlation Structure

The one-point statistics of \( E, \sigma_{\max}, \varepsilon_{\max} \), and \( TEA \) are shown in Fig. 21 in a probability paper format with a logarithmic axis for the cumulative probability set in such a way as to result in a straight fit through the data points should they be Gaussian. While these data show only small departures from Gaussianity, Beta turns out to be the probability distribution of the best overall goodness-of-fit for a wide range of specimen sizes and both origins of paper (Champion and Repap). Beta is shown as a broken line. Its density function, for a random variable \( x \), is given by

\[
f(x; a_1, a_2, \delta_1, \delta_2) = \left[ \left( \frac{x - \delta_1}{\delta_2 - \delta_1} \right)^{a_1 - 1} \left( 1 - \frac{x - \delta_1}{\delta_2 - \delta_1} \right)^{a_2 - 1} \right] / [(\delta_2 - \delta_1)B(a_1, a_2)] \quad (4.3)
\]

where \( a_1, a_2, \delta_1, \) and \( \delta_2 \) are adjustable parameters, and \( B(a_1, a_2) = \Gamma(x)\Gamma(y)/(\Gamma(x+y)) \) is the so-called beta function.

A number of other distributions were tried---in particular, Chi, Chi-Square, Exponential, Fréchet, Gumbel Min, Log-Normal, Rayleigh, and Weibull. However, none of them offered the same overall universality combined with the goodness-of-fit as beta. Interestingly, the same type of observation was recently made in the context of elasticity and damage micromechanics of disordered two-phase composites with microscale elastic-brittle response [36, 37].

The means of \( E, \sigma_{\max}, \varepsilon_{\max} \), and \( TEA \) are, respectively, 3980, 34.9, 1.9, and 0.46, while their coefficients of variation are 0.04, 0.05, 0.1 and 0.14. It is most interesting to note here that the COV of \( E \) is on the same order as that of \( \sigma_{\max} \).
Fig. 21. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 20.

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The fundamental two-point, second-order information on the relation of two components \( u_i \) and \( u_j \) of the underlying vector field \( \hat{u} \) is specified by a correlation coefficient

\[
\rho_{ij}(x_1, x_2) = \frac{\langle C_i(x_1)C_j(x_2) \rangle - \langle C_i(x_1) \rangle \langle C_j(x_2) \rangle}{\sigma_i \sigma_j}
\] (4.4)

which takes values between 1 and -1. These two extreme values are called, respectively, full positive and full negative correlatedness, while \( \rho_{ij} = 0 \) represents zero-correlatedness. The case \( i = j \) is termed the auto-correlation, while \( \rho_{12}, \rho_{13}, \ldots \), are called cross-correlation.

There are, clearly, four auto-correlations and six cross-correlations, all of which can be put into a symmetric \( 4 \times 4 \) array whose lower tridiagonal only is shown below (recall definition (4.2))

\[
\rho_{ij}(x_1, x_2) = \begin{bmatrix}
\rho_{E,E} & \rho_{E,\sigma_{\max}} & \rho_{E,\varepsilon_{\max}} & \rho_{E,TEA} \\
\rho_{E,\sigma_{\max}} & \rho_{\sigma_{\max}\sigma_{\max}} & \rho_{\sigma_{\max}\varepsilon_{\max}} & \rho_{\sigma_{\max}TEA} \\
\rho_{E,\varepsilon_{\max}} & \rho_{\sigma_{\max}\varepsilon_{\max}} & \rho_{\varepsilon_{\max}\varepsilon_{\max}} & \rho_{\varepsilon_{\max}TEA} \\
\rho_{E,TEA} & \rho_{\sigma_{\max}TEA} & \rho_{\varepsilon_{\max}TEA} & \rho_{TEA,TEA}
\end{bmatrix}
\] (4.5)

Thus, when we consider correlations between the nearest neighbors (contiguous specimens)---i.e., when \( x_2 = x_1 \pm L \)---we cannot expect the diagonal entries to equal unity. This is the setup which begins to shed light on the spatial correlation structure. We can do it with our data at hand (Fig. 20) providing we assume a wide-sense stationarity of the \( \hat{u} \) field, that is

\[
\rho_{ij}(x_1, x_2) = \rho_{ij}(\ell) \quad \text{for} \quad \forall \ell = x_1 - x_2
\] (4.6)

Additionally, we need to assume a space-ergodic property, that is, an interchangeability of the ensemble \( \langle \langle \ \rangle \rangle \) with the spatial averages \( \langle \rangle \)

\[
\langle u_i(x, \omega) \rangle = \overline{u_i(x, \omega)} \quad \text{for} \quad \forall x \quad \text{and} \quad \forall \omega \in \Omega
\] (4.7)

Strictly speaking, we are working here with quasi-stationary and quasi-ergodic fields at best, but this Bayesian point of view is sufficient to overcome the epistemological dilemma of a human inability to predict mechanical (and other) properties of paper with an absolute certainty; see e.g., [2]. These issues are only briefly signalled here for completeness of this presentation---their full analysis is being conducted under our NSF grant.
The $\rho_{ij}$'s for the nearest neighbors in the machine ($x$) and cross ($y$) directions are

$$
\rho_{ij}(L_x) = \begin{bmatrix}
0.24 \\
0.21 & 0.27 \\
0.00 & 0.10 & 0.21 \\
0.14 & 0.23 & 0.15 & 0.22
\end{bmatrix} \\
\rho_{ij}(L_y) = \begin{bmatrix}
0.33 \\
0.18 & 0.13 \\
0.09 & 0.02 & 0.26 \\
0.05 & 0.04 & 0.04 & 0.04
\end{bmatrix}
$$

(4.8)

The actual numbers in (4.6) depend on various factors---type of paper, specimen size and shape, fiber, fiber-fiber bonding, floc and streak structure, etc.---but it is already apparent that $\tilde{u}$ is a quasi-isotropic random field (e.g., [34]).

Next, focusing on the $\rho_{ij}$ functions at a point---i.e., when $x_1 = x_2$---we find

$$
\rho_{ij} = \begin{bmatrix}
1 \\
\rho_{12} & 1 \\
\rho_{13} & \rho_{23} & 1 \\
\rho_{14} & \rho_{24} & \rho_{34} & 1
\end{bmatrix}
$$

(4.9)

where $\rho_{ij} = \rho_{ij}(x_1, x_1)$. From this we observe that:

i) Cross-correlations between $E$ and inelastic parameters $\sigma_{max}^i, \epsilon_{max}^i$, and $TEA$ are weak, although we note that $\rho_E, \sigma_{max}^i$ is greater than $\rho_E, \epsilon_{max}^i$ or $\rho_E, TEA$.

ii) Three cross-correlations between $\sigma_{max}^i, \epsilon_{max}^i$, and $TEA$ are about the same.

The analysis reported in this and the previous section (i.e., 4.2 and 4.3) also appears in [38].

### 4.4 Grey-scale plots for other specimen sizes (Champion paper)

This section collects the grey-scale plots of all other specimen sizes for the Champion paper, as per Table 3.
Fig. 22. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{\text{max}}$ in lbf/in; (c) strain to failure $\varepsilon_{\text{max}}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $2 \times 100$ array of $1'' \times 1/8''$ specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 23. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\epsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a 20 x 20 array of 1” x 1” specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 24. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $2 \times 100$ array of $7'' \times 1/8''$ specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 25. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\epsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a 25 x 4 array of 7" x 10" specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 26. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{\text{max}}$ in lbf/in; (c) strain to failure $\epsilon_{\text{max}}$ in $\%$; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $4 \times 25$ array of $28'' \times 1''$ specimens. The ranges and assignments of values are shown in the respective insets.

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Fig. 27. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{\text{max}}$ in lbf/in; (c) strain to failure $\varepsilon_{\text{max}}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $10 \times 10$ array of $28'' \times 4''$ specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 28. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in $\%$; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $25 \times 4$ array of $28'' \times 10''$ specimens. The ranges and assignments of values are shown in the respective insets.

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Fig. 29. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $30 \times 1$ array of $28'' \times 79''$ specimens. The ranges and assignments of values are shown in the respective insets.

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4.5 Empirical histograms and Beta probability distribution fits (Champion paper)

Fig. 30. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 22.

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Fig. 31. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 23.
Fig. 32. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 24.

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Fig. 33. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 25.
Fig. 34. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 26.
Fig. 35. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 27.
Fig. 36. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 28.
Fig. 37. Top to bottom: empirical histograms and Beta probability distribution fits of four mechanical properties shown in Fig. 29.
4.6 Grey-scale plots for all specimen sizes (Repap paper)

This section collects the grey-scale plots of all specimen sizes for the Repap paper, as per Table 2. The empirical histograms and Beta probability distribution fits were given in the last year's PAC report in Figs. 10-16.
Fig. 38. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $2 \times 100$ array of $1'' \times 1/8''$ specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 39. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbm/in; (c) strain to failure $\epsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $25 \times 8$ array of 1" x 1" specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 40. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{\text{max}}$ in lbf/in; (c) strain to failure $\varepsilon_{\text{max}}$ in $\%$; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $2 \times 100$ array of $7'' \times 1/8''$ specimens. The ranges and assignments of values are shown in the respective insets.

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Fig. 41. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a 10 x 20 array of 7" x 1" specimens. The ranges and assignments of values are shown in the respective insets.

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Fig. 42. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $50 \times 2$ array of $7'' \times 10''$ specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 43. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $5 \times 20$ array of 28”×1” specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 44. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ in lbf/in; (c) strain to failure $\varepsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a $20 \times 5$ array of 28" $\times$ 4" specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 45. A grey-scale plot of: (a) elastic modulus $E$ lbf/in; (b) breaking strength $\sigma_{max}$ lbf/in; (c) strain to failure $\epsilon_{max}$ in %; and (d) tensile energy absorption $TEA$ lbf/in. All data are for a 50 x 2 array of 28" x 10" specimens. The ranges and assignments of values are shown in the respective insets.
Fig. 46. A grey-scale plot of: (a) elastic modulus $E \text{ lbf/in}$; (b) breaking strength $\sigma_{\text{max}}$ in $\text{lbf/in}$; (c) strain to failure $\epsilon_{\text{max}}$ in $\%$; and (d) tensile energy absorption $TEA \text{ lbf/in}$. All data are for a $30 \times 1$ array of $28'' \times 40''$ specimens. The ranges and assignments of values are shown in the respective insets.
4.7 Conclusions

A detailed description of all the tests and further data are given in []. Here we give the principal conclusions regarding the variability and cross-correlations of $E$, $\sigma_{\text{max}}$, $\varepsilon_{\text{max}}$, and $\text{TEA}$. In general, the data shows that $E$ increases with increasing length at a constant width and decreases with increasing width at a constant length. $E$ increases with aspect ratio from 0.35 to 7, but then remains unchanged when the aspect ratio is raised to 56. For the most part, $\sigma_{\text{max}}$ decreases with increasing length, but shows no clear relationship with increasing width at a given length or aspect ratio. $\varepsilon_{\text{max}}$ decreases with increasing length and shows no correlation with increasing width or aspect ratio. $\text{TEA}$ decreases with increasing length at a constant width and shows no clear trend with increasing width or aspect ratio. Overall, changing the sample length has a more pronounced and clearer effect on a tensile property than changing the width.

The scatter in $E$ decreases with increasing length, but there is no dependence on width. The scatter in $\sigma_{\text{max}}$ increases with increasing length and width, although the width effect is minimal. Strain data indicates a reduction in variation with increasing length, and no definite relationship between variation and increasing width. Variation in $\text{TEA}$ decreases, in general, with increasing length and increases with increasing width. Many of the samples contradict the weak link theory showing increasing variation with increasing dimension. None of the four properties showed any change in variation with a change in aspect ratio.

The one point-correlation analysis of the tensile properties shows no relationship between $E$ and $\varepsilon_{\text{max}}$ or $\text{TEA}$. There is a very weak correlation, around 0.5, between $E$ and $\sigma_{\text{max}}$ for about half of the data sets, but no correlation is found or, actually, negative correlations are found in the remaining half of the data sets. The $\sigma_{\text{max}}$, $\varepsilon_{\text{max}}$, and $\text{TEA}$ correlate strongly, quite often over 0.9 with each other. Of the eighteen data sets, the correlation coefficient between $\sigma_{\text{max}}$ and $\text{TEA}$ is the highest in two thirds of them. With the exception of the 28x79 data set, the $\sigma_{\text{max}}$-$\text{TEA}$ correlation is the strongest in all of the Champion data sets. The correlation between $\varepsilon_{\text{max}}$ and $\text{TEA}$ is the strongest correlation for five of the remaining data sets. The $\sigma_{\text{max}}$-$\varepsilon_{\text{max}}$ correlation is the highest for the 7x1/8 Repap data set. In general, the strongest correlation is between $\sigma_{\text{max}}$ and $\text{TEA}$ with an average of 0.7, followed by the correlation between $\sigma_{\text{max}}$ and $\varepsilon_{\text{max}}$ which is significantly lower at an average of ~0.6. The average correlation coefficient between $E$ and stress is 0.32, between $E$ and $\varepsilon_{\text{max}}$ ~0.16, and between modulus and $\text{TEA}$ ~0.06.
4.8 Further Directions

1. The basic model of a single fiber (treated as a helically wound composite) coupling its extensional with the torsional responses, will be advanced to account for:
   - hygroscopic effects;
   - dependence of the fibril angle on the radial distance from the fiber's axis;
   - presence of more than one layer of fibrils;
   - shear and bending response;
   - a non-circular (i.e., squashed rectangular) cross-section.

2. The computational mechanics model of complex 3-D networks of thousands of fibers will be extended to:
   - include inelastic micro-strains so as to model elastic-inelastic-fracture phenomena;
   - include dynamic effects, so as to assess the dispersion and damping relations in stress wave propagation in paper for a wide range of frequencies;
   - networks larger by a significant factor so as to simulate systems of hundreds of thousands of fibers.

3. The computational mechanics model's predictions will be further verified by a comparison to deformation patterns and measured mechanical properties of low basis weight paper.

4. The multiscale statistical model of scale effects of effective mechanical responses in the plane of the web will be further developed. This will include a continuation of uniaxial tests in MD, as well as new uniaxial tests in CD and new biaxial tests.

5. Detailed optical observations of damage/crack evolution and its correlation with fiber network structure and formation will be carried out.
5. Significant Findings and Conclusions to Date

1. A basic model of a single fiber treated as a helically wound composite has been formulated. The model reveals the existence of two waves (fast and slow) coupling the extensional with the torsional responses of the fiber.

2. A computational mechanics program for study of elasticity as well as progressive damage of complex 3-D networks of thousands of fibers, rigorously accounting for a multitude of microscale interactions and formation (flocculation) structure, has been developed.

3. Model’s predictions are well verified by a comparison to deformation patterns observed on low-density paper.

4. A general framework of experimentally observed scale effects of effective mechanical responses in the plane of the web, along with the associated statistics and the correlation structure, has been developed. This framework employs a multiscale model, which is motivated by the fact that the paper structure can be regarded as a frozen-in turbulence of fiber suspensions.

6. Commercial Impact

1. Savings of raw materials for same or better product performance (mechanical and transport properties) through prediction and optimization of fiber network structures. Tool: a user-friendly computer software for simulation of mechanical interactions in realistic networks of thousands of fiber-beams.

2. Optimization of paper making processes and paper products through characterization of statistical and scale-dependent elastic and strength properties of paper webs.
BIBLIOGRAPHY


[29] G. Peter (1999), *Private communication*, IPST.


IMPROVING THE REFINING OF CHEMICAL PULPS

STATUS REPORT

FOR

PROJECT F024

John Waterhouse
Hiroki Nanko
Miranda Bliss

Institute of Paper Science and Technology
500 10th Street, N. W.
Atlanta, Georgia 30318
DUES FUNDED PROJECT SUMMARY

Project Title: Improving the Refining of Chemical Pulps
Project Code: PROPTY
Project Number: F024
PAC: Paper Physics
Division: Fiber and Paper Physics
Project Staff:
   Faculty/Senior Staff: John F. Waterhouse, Hiroki Nanko
   Staff: Miranda Bliss
FY 98-99 Project Budget: $ 89,000
   Allocated as Matching Funds: none
Time Allocation
   Faculty/Senior Staff: Waterhouse 25%, Nanko 10%
   Staff: Bliss 50%
Supporting Research
   M.S. Students: None
   Ph.D. Students: None
   Related External Funding: None

RESEARCH LINE/ROAD MAP:

10. Reduce net energy consumption per ton by 30% compared to '97' levels.
   - reduce energy consumption

11. Convertability and End-Use Performance

Improve the ratio of product performance to cost for pulp and paper products by 25% by developing:
   - models, algorithms, and functional samples of fibrous structures and coatings, which describe and demonstrate improved convertibility and end use performance, and breakthrough papermaking and coating processes which can produce innovative webs with greater uniformity than achieved by current processes

PROJECT OBJECTIVE:

Define and determine a pulp's refinability behavior, and how changes in fiber structure, produced by refining, are related to an improved balance between paper machine runnability, i.e. water removal, and paper properties, i.e., strength improvement.
PROJECT BACKGROUND
This project was initiated in October 1996. Its value to member companies includes: 1) improved utilization of raw materials, 2) increases in paper-machine productivity, 3) improved tools for pulp characterization, 4) enhanced control of refining process, and 5) energy reduction associated with refining and water removal.

The main project goal is to develop a pulp refinability index, i.e., its propensity to cut, fibrillate, produce fines, and to curl and microcompress, which can be applied to pulps produced by new pulping and bleaching processes, genetically modified pulps, as well as pulps produced by more conventional pulping and bleaching methods.

A concomittant goal is to produce and characterize specific changes in fiber structure and measure their impact on water removal and paper properties. The specific changes in fiber structure are: 1) fines production, 2) external changes in fiber structure, 3) internal changes in fiber structure, and 4) curl removal.

Ultimately, we would like to determine for both laboratory and production refiners the extent to which these changes are produced with selected furnishes and the means for controlling them.

SUMMARY OF RESULTS (March 98 - March 99)

1) Joint project with CBSD to examine refining behavior of different yield pulps and bleaching sequences – characterization and refining of the pulps, as well as handsheet making and the measurement of physical properties, is almost complete.
2) Implementation of Pd and Fe staining techniques for measurement of internal changes, external changes, and secondary fines indices.
3) Examination of cross sections of Fe colloid stained PFI refined fibers using the transmission electron microscope.
4) Exploratory experiments to produce specific changes in fiber structure.

PROJECT GOALS FOR FY 99-00

1. Develop methods to produce specific changes in fiber structure by refining and the means for their characterization.

2. Determine the extent to which specific changes in fiber structure is produced in production refining systems as a function of C factor.
3. In conjunction with goals 1. and 2. measure wet and dry state properties as they might relate to machine runnability and paper performance.

PROJECT DELIVERABLES

1. Methodology for determining a pulp's response to refining.


3. Strategies for reducing energy consumption and/or improving paper machine productivity.

PROJECT SCHEDULE

March 99 - March 00

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INTRODUCTION

Refining or beating (terms used interchangeably) is defined as the changes in fiber structure necessary to maximize the papermaking potential of a pulp. Papermaking potential is concerned with achieving the right balance between important paper properties and machine runnability within specified economic restraints.

We have previously identified, from the review by Page [1], 9 changes in fiber structure produced by refining. In addition, we have chosen to examine more closely the contribution of the changes given below.

1. Internal changes in fiber structure
2. External changes in fiber structure
3. Fines production
4. Changes in fiber curl
5. Fiber length reduction

The major tasks identified at our Project Advisory Committee Meeting on March 26, 1997 are as follows:

- Select and characterize pulp types to be used.
- Develop methods to produce specific refining actions.
- Measure wet and dry state properties as they might relate to machine runnability and paper performance.

Following discussions with RAC April 1998, an attempt was made to give more focus to the project and resulted in the following short term objectives:

1. Establish methods for producing selected changes in fiber structure
2. Establish methods to measure and characterize these changes in fiber structure.
3. Determine the relative impact of these changes on drainage, water removal, and paper properties.
4. Determine how selected pulp variables influence these changes in fiber structure.
5. Determine the extent to which these changes can be produced in production refiners.

Select and characterize pulp types to be used – Joint CBSD-FPPD Project

It was agreed that it would be of mutual benefit to cooperate with the Chemical and Biological Sciences Division (CBSD) in obtaining pulps for this project. The CBSD is ultimately interested in improved pulping and chlorine-free bleaching
strategies. It is interesting to note that of the approximately 855 categories of tasks and measurements 83% are complete as of this writing.

The pulps are identified in Table 1. The species is loblolly pine and the kappa number ranges from a high of 110 to a low of 10.7 for the unbleached pulps, which have been delignified using either a conventional kraft cook or a kraft cook to kappa number 30 followed by oxygen delignification. The final kappa number and pulp brightness for the CED and DED bleaching stages are also included in Table 1. The carboxyl content of these pulps was also determined and the results are given in Table 2.

Table 1. Kappa Number and Brightness of Pulps Prepared by the CBSD

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. kappa no.</th>
<th>CED kappa no.</th>
<th>R_∞</th>
<th>DED kappa no.</th>
<th>R_∞</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>0.87</td>
<td>84.0</td>
<td>2.10</td>
<td>75.9</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>0.85</td>
<td>86.9</td>
<td>1.28</td>
<td>84.7</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>3.51</td>
<td>88.3</td>
<td>6.28</td>
<td>81.2</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>2.80</td>
<td>88.6</td>
<td>1.31</td>
<td>83.1</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>0.46</td>
<td>90.8</td>
<td>1.03</td>
<td>84.1</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>1.11</td>
<td>89.9</td>
<td>4.58</td>
<td>85.8</td>
</tr>
</tbody>
</table>

S.P. chips - refers to southern pine chips

Table 2. Carboxyl Content of Pulps Prepared by the CBSD

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. kappa no.</th>
<th>Unbl. COOH meq/g</th>
<th>Bl. CED COOH meq/g</th>
<th>Bl. DED COOH meq/g</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>0.175</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>0.077</td>
<td>0.030</td>
<td>0.041</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>0.049</td>
<td>0.037</td>
<td>0.039</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>0.081</td>
<td>0.046</td>
<td>0.041</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>0.078</td>
<td>0.036</td>
<td>0.046</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>0.072</td>
<td>0.051</td>
<td>0.035</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>0.067</td>
<td>0.044</td>
<td>0.043</td>
</tr>
</tbody>
</table>

The conductivity, pH and ionic nature of the pulps can affect their refinability [2,3], and, therefore; we need to be consistent in preparing the pulps for this purpose. After washing, the pulps were converted to the calcium form. The

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outline of the procedure is as follows. The pulps were washed 3 times in deionized water and then the pH was reduced to 1.5 using HCl. The pulps were then washed in nano water, and then soaked in a 0.1M CaCl solution. The pH was adjusted to 9.5 using CaOH. Following this, the pulps were washed and soaked in nano water until the pH was approximately 7. After conversion to the calcium form, a metals analysis was performed. The calcium content of the pulps is shown in Table 3.

Table 3. Final Calcium Content of Pulps Prepared by the CBSD

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. kappa no.</th>
<th>Unbl. mg/kg</th>
<th>Bl. CED mg/kg</th>
<th>Bl. DED mg/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>4360</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>1680</td>
<td>907</td>
<td>1290</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>903</td>
<td>647</td>
<td>764</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>1720</td>
<td>663</td>
<td>755</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>1610</td>
<td>614</td>
<td>745</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>1430</td>
<td>706</td>
<td>740</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>1420</td>
<td>700</td>
<td>717</td>
</tr>
</tbody>
</table>

Refining of the above pulps was carried out in IPST's PFI mill at 3000 revolutions. Characterization methods include Canadian Standard Freeness, fines content (material passing through a 200 mesh screen using the Britt jar technique), fiber length and curl, water retention value (WRV whole and fines free pulp), Fe and Pd staining, and selected paper properties including, zero span, scattering and absorption coefficient, gurley porosity, elastic (ultrasonic in-plane and out-of-plane), and failure properties. Initially, our intention is to evaluate the unrefined pulp and pulp beaten to 3000 revolutions in the PFI mill using 24 g/o.d. pulp at 10% consistency (i.e., the CPPA method). As methods for selected refining action become available, these will also be used.

Page [4] has stated that the tear-tensile relationship should be an excellent method for monitoring the effects of pulp delignification. It is not expected that we will be able to examine in detail all of the above pulps. Initially, we will investigate the effects of kappa number change as a result of conventional kraft pulping and combined kraft and oxygen delignification as shown in Table 1.
Develop methods to produce specific refining actions

Refining action is central to the structural changes produced in the fiber. C Factor analysis developed by Kerekes et al. [5–7]) is one approach to defining refining action in terms of the number and severity of impacts a fiber receives in the refiner. Changes in fiber structure are given at two extremes, i.e., low severity with a high number of impacts produces "fibrillation", while the converse high severity and a low number of impacts produces "cutting". What happens between these extremes is largely unknown. The following diagram indicates one possibility.

![Diagram]

Internal Changes in Fiber Structure

It has been suggested [8,9] that internal changes in fiber structure are mainly created through a fatigue process. In this regard, we are examining more carefully what happens in the PFI mill including the early experiments of Stephansen [10] in the development of the PFI mill and the more recent work of Kerekes's group.

External Changes in Fiber Structure

A number of schemes involving abrasive particles and surfaces are under consideration.

Fines Production

It is hypothesized that the secondary fines produced by refining and external changes in fiber structure go hand in hand, although in the present situation the relative amounts of each may well be kappa number dependent. In any case fines will be produced in either the PFI mill or valley beater. A homogenizing
device is also available which may be satisfactory for generating fines from small quantities of pulp.

The outer secondary wall $S_1$ of both earlywood and latewood fibers is probably the main source of fines during the early stages of refining [11]. It has been hypothesized that the production of fines is initially quite rapid but slows down when fines are predominantly produced from the middle secondary wall or $S_2$ layer [12]. We do not know what the relative rate of earlywood and latewood fines production might be or their impact on water removal and paper properties.

Changes in Fiber Curl

Curl (kinks, microcompressions) can have a strong impact on many pulp and paper properties. It is relatively easy to induce fiber curl, e.g., by high consistency processing; however, its removal is more problematic. Low consistency processing, i.e., < 1.5% consistency, may be effective for curl removal, but not necessarily practical. Nevertheless, we need to know how effective curl removal is on selected properties.

We are in the process of determining the pulp's potential as a function of kappa number and bleaching conditions. It is not anticipated that the PFI refiner will induce significant curl and, if anything, should reduce any curl present.

In the results section we shall see that the oxygen stage of delignification, which was performed with a peg mixer at around 10% consistency, has resulted in a significant level of curl being produced.

Fiber Length Reduction

One of the major differences between laboratory refiners, e.g., PFI mill and production refiners, e.g., disk refiners, is that the action of the former is both gentle and homogeneous while that of the latter can be severe and heterogeneous. The latter may result in excessive fines production as well as a significant reduction in fiber length.

Fiber length reduction may result from a cutting or shear action but perhaps more likely from the tensile stresses that the fibers are subjected to when passing between the bars in a refiner. Therefore, we hypothesize that the strength of the fibers in the never-dried wet state should be an indication of a fiber's propensity to cutting.

We had intended to carry out experiments in the Escher–Wyss laboratory refiner to either prove or disprove this hypothesis. However, circumstances have been
such that we have not yet been able to conduct more realistic or production like refining experiments.

Kerekes [13] has shown see diagram below that the severity of impacts in a refiner is such that as a result of fiber cutting, tensile strength is reduced at a constant specific energy input.

The above diagram is mainly based on literature data and it appears that for some reason hardwood fibers are being treated more severely than they should, i.e., they already have a lower fiber length than softwoods. Interestingly, there are two regions at low severity. Region A would be highly desirable while region B is to be avoided. The conditions for landing in either of these regions are presently unknown.

**Measure wet and dry state properties as they might relate to machine runnability and paper performance**

Established methods will be used where possible to monitor pulp and paper properties both with conventional refining, i.e., the PFI mill, and to assess the contribution of the specific refining actions discussed above. These include CSF, WRV, and surface area measurements in both the swollen and dried state, utilizing both whole and fines free pulps. Pd and Fe staining and microscopy are examples of newer tools to be used in assessing the changes in fiber structure produced by a specific refining action. Zero span measurements will be used in the never-dried, dried, and rewet conditions to measure fiber strength.
RESULTS AND DISCUSSION

Select and characterize pulp types to be used

As shown in Tables 1, 2, and 3 a set of unbleached pulps have been produced by the CBSD using kraft, and kraft plus oxygen delignification stages. The two bleaching-stage sequences, i.e., CED and DED, result in further delignification and an increase in pulp brightness. We note that CED generally results in a lower kappa number and higher brightness than the DED sequence.

For the unbleached pulps, carboxyl content falls as kappa number falls, but at a given kappa number kraft plus oxygen delignification yields a pulp with a higher carboxyl content. The CED and DED bleaching sequences result in further carboxyl content reduction.

As delignification proceeds, calcium content for the unbleached pulps is also reduced, and the pattern of variation is similar to that found for carboxyl content. Generally, the calcium content of the CED sequence pulps is lower than the DED sequence pulps.

Measure wet and dry state properties as they might relate to machine runnability and paper performance

Wet State Properties

CSF, fines content, WRV's, Fe and Pd staining results are shown in Tables 4 through 10.

It is acknowledged that the PFI mill does not simulate production refining conditions. The action is such that it does not approach the intensity or severity of a disk refiner. Thus a pulp's propensity to cutting is unlikely to be revealed when treated in a PFI mill. Nevertheless, the refiner does produce internal and external changes in fiber structure and concomitantly generates fines. In addition, curl reduction can also occur. These changes are the result of cyclic stresses being applied to the fibrous mat as well as abrasive action between the fibers.

Presumably, the extent to which changes in fiber structure take place will be determined by such factors as 1) pulping and bleaching conditions, 2) species and morphology, 3) pulp rheology, 4) consistency and state of flocculation, 5) inter-fiber friction, 6) pH and ionic state, 7) fiber deformation behavior, and 8) chemical composition of the cell wall. Clearly some of these factors are interrelated.

It is interesting to note that freeness reduction, in refining from 0 to 3000 revolutions increases with decreasing kappa number (see Tables 4a and 4b).
similar trend is also found with the CED and DED bleaching sequences. The level of freeness change is about the same as that found for the unbleached pulps. Thus, the freeness change appears to be controlled by the initial delignification step and is less influenced by bleaching.

There is no clear correlation between the amount of fines generated by refining as kappa number is decreased (see Tables 5a and 5b). For the unbleached pulp the overall average fines produced is 5.97% with a variation of 10.4%. The overall averages and variability for the CED and DED sequences, respectively, are 6.47%, and 10.8%, and 6.26%, and 16.5%. Since the expected increase in CSF does not appear to be associated with a significant variability in fines content, it is possible that the hydrodynamic surface area of the fines increased with decreasing kappa number.

The water retention value (WRV) results are summarized in Tables 6 and 7 for the whole and fines free pulps, respectively. Changes in WRV with refining are given in Table 8 for the whole and fines free pulps. We note that the water uptake for the kraft-oxygen delignified pulps is about twice as high as the kraft only pulps. This is attributed to low WRV's of the unrefined kraft-oxygen delignified pulps and subsequently greater swelling of the cell wall. The reason for the low unrefined values is not yet known.

The iron and palladium colloid measurements made on the fines-free fraction of the unrefined and refined pulps are shown in Tables 9a, 9b, 10a, and 10b. The iron colloid is small and is thus capable of penetrating cracks in the cell wall. By comparison, the palladium colloid is large and not capable of penetrating the cell wall and, therefore, should be sensitive to the external surface of the fibers.

The Fe and Pd results for bleach sequences CED and DED are still incomplete. The Fe colloid results for the unbleached pulp do not indicate a clear trend or difference between the two methods of delignification, although the uptake of iron colloid increases with refining. In view of the WRV's shown in Table 8 for the two methods of delignification, we might have expected a similar difference in the Fe colloid measurements. The Pd colloid staining results clearly show an increase with refining but the trend with type and amount of delignification is not clear.

**Fiber Properties**

Fiber properties measured include fiber length distribution (Kajaani FS-100), fiber curl (projection method), and zero span (Pulmac zero span tester), as summarized in Tables 11 to 15. This data set is incomplete.

There is a small reduction in weighted/weighted average fiber length with refining for the kraft pulps. However, this seems to be consistent with a small increase in fiber curl as shown in Table 12. On the other hand, the two kraft plus oxygen
delignified pulps show an increase in weighted/weighted average fiber length. Again, if we examine the curl results, we see that the initial curl of these fibers is quite large, i.e., 1.42 and 1.45, however, PFI refining at 3000 revolutions was not able to completely remove all of the curl.

We have experienced significant problems with our zero span tester resulting in its return to the manufacturer twice for service. We are in the process of redoing our zero span results since the initial set is considered to be unreliable. The results, albeit incomplete, are shown in Tables 13, through 18 for never-dried (wet), dried, and dried and re-wet samples. It can be seen that delignification and bleaching can result in a significant loss in fiber strength especially when the wet never dried state is compared with the dried state, i.e., the wet/dry ratios for DED initial kappa's of 28.1 and 10.7 are 0.800 and 0.554, respectively. Again it is hypothesized that a reduction in the wet to dry strength ratio may be an indication that the pulp has a greater propensity to cutting. It is also interesting to note that refining appears to further decrease this ratio. On the other hand the once dried ratio is not as low as the never-dried ratio perhaps indicating that "hornification" and attendant cross-linking is responsible for the reduced loss.

Handsheet Properties

Handsheet physical and strength related properties are summarized in Tables 19 and 20 for the unrefined and refined pulps, respectively. A detailed commentary will be reserved until the data set is complete, particularly in the absence of a complete set of zero span data.

We compare, initially, the performance of the unbleached kraft and unbleached kraft plus oxygen delignification refined pulps shown in Table 19. We note that oxygen treated pulps densify to a much greater extent than the kraft pulps and this is possibly attributed to the greater degree of swelling of those pulps (see Table 8). On balance, the elastic and strength properties are higher for the kraft pulp, while the tear performance for the oxygen delignified pulp is higher in spite of the higher sheet density. The reason for the increased tear performance may possibly be attributed to some residual curl remaining even after refining.

The variation of tear and tensile indices with kappa number for the unrefined and refined pulps is shown in Figures 1 through 4. In the unbeaten case, both tear and tensile values are significantly higher for the kraft and bleached kraft pulps. The kraft and bleached kraft pulps appear to give a superior performance for tensile index whereas the kraft-oxygen delignified pulps yield a superior tear performance.

Conclusions

Since the data set is incomplete, it is not possible to draw any strong conclusions at this time with respect to refining. However, it is clear that the process of
delignification has a strong influence on pulp and paper properties, perhaps, mainly due to chemical differences, e.g., kappa number, method of delignification and bleaching sequence.

In order to have the same basis for comparison we put all pulps into the same ionic form, i.e., Ca. However, the amount of Ca varies with kappa number and roughly parallels the variation in carboxyl content and this may affect refining action.

With respect to the changes in fiber structure we have selected, it is not yet clear how internal and external changes are influenced by the variation in pulp treatment. The influence on fines production appears to be quite small, although there are some exceptions. Changes in fiber length appear to parallel changes in fiber curl, and thus not significant reduction in fiber length is found. On the other hand, from limited results fiber strength, particularly never dried strength appears to be significantly affected by the pulping process, and, therefore, we would expect a much different response in a production refining situation, i.e., the likelihood of fiber "cutting". The latter remains a hypothesis to be verified in future work.

LITERATURE CITED


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Acknowledgments

The author would like to thank Miranda Bliss for her painstaking and careful work to date on this project and also to Barbara Lee Hing for her editorial revision.
## Table 4a. CSF Values of Whole Pulps Refined in the PFI Mill

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl.</th>
<th>CED</th>
<th>DED</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>CSF ml 0 revs</td>
<td>CSF ml 3000 revs</td>
<td>CSF ml 0 revs</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>751</td>
<td>669</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>743</td>
<td>400</td>
<td>723</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>727</td>
<td>342</td>
<td>738</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>753</td>
<td>364</td>
<td>728</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>740</td>
<td>338</td>
<td>733</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>732</td>
<td>296</td>
<td>730</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>755</td>
<td>347</td>
<td>740</td>
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</table>

## Table 4b. ΔCSF Values of Whole Pulps Refined in the PFI Mill

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl.</th>
<th>CED</th>
<th>DED</th>
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</thead>
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<td></td>
<td>ΔCSF ml 0 - 3000 revs</td>
<td>ΔCSF ml 0 - 3000 revs</td>
<td>ΔCSF ml 0 - 3000 revs</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>82</td>
<td>-</td>
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<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>343</td>
<td>317</td>
<td>350</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>385</td>
<td>358</td>
<td>358</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>389</td>
<td>387</td>
<td>362</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>402</td>
<td>413</td>
<td>378</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>436</td>
<td>452</td>
<td>395</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>408</td>
<td>447</td>
<td>492</td>
</tr>
</tbody>
</table>

## Table 5a. % Fines of Pulps Refined in the PFI Mill

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl.</th>
<th>CED</th>
<th>DED</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0 revs</td>
<td>3000 revs</td>
<td>0 revs</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>1.28</td>
<td>7.64</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>1.80</td>
<td>7.30</td>
<td>1.81</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>2.52</td>
<td>9.22</td>
<td>2.19</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>2.52</td>
<td>7.36</td>
<td>1.72</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>2.03</td>
<td>7.96</td>
<td>1.18</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>2.03</td>
<td>8.25</td>
<td>1.76</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>1.36</td>
<td>7.57</td>
<td>1.99</td>
</tr>
</tbody>
</table>
Table 5b. Δ% Fines Values of Pulps Refined in the PFI Mill

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl. ΔCSF ml 0 - 3000 revs</th>
<th>CED ΔCSF ml 0 - 3000 revs</th>
<th>DED ΔCSF ml 0 - 3000 revs</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>6.36</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>5.50</td>
<td>5.86</td>
<td>5.92</td>
</tr>
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<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>6.70</td>
<td>6.13</td>
<td>5.47</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>4.84</td>
<td>5.62</td>
<td>6.85</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>5.93</td>
<td>7.39</td>
<td>5.81</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>6.22</td>
<td>6.94</td>
<td>5.44</td>
</tr>
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<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>6.21</td>
<td>6.86</td>
<td>8.09</td>
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</table>

Table 6. WRV of Whole Pulps Refined in the PFI Mill

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl. 0 revs 3000 revs</th>
<th>CED 0 revs 3000 revs</th>
<th>DED 0 revs 3000 revs</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>1.70 2.17</td>
<td>- -</td>
<td>- -</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>1.83 2.41</td>
<td>1.80 2.34</td>
<td>1.73 2.39</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>1.87 2.33</td>
<td>1.70 2.19</td>
<td>1.68 2.22</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>1.53 2.31</td>
<td>1.60 2.27</td>
<td>1.64 2.32</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>1.55 2.32</td>
<td>1.60 2.30</td>
<td>1.61 2.23</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>1.59 2.37</td>
<td>1.60 2.28</td>
<td>1.61 2.18</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>1.55 2.27</td>
<td>1.62 2.29</td>
<td>1.63 2.32</td>
</tr>
</tbody>
</table>

Table 7. WRV of Fines-Free Pulps

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl. 0 revs 3000 revs</th>
<th>CED 0 revs 3000 revs</th>
<th>DED 0 revs 3000 revs</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>1.74 2.00</td>
<td>- -</td>
<td>- -</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>1.84 2.07</td>
<td>1.72 2.21</td>
<td>1.70 2.16</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>1.89 2.11</td>
<td>1.64 1.93</td>
<td>1.63 1.96</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>1.54 2.00</td>
<td>1.57 1.99</td>
<td>1.54 2.03</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>1.54 2.03</td>
<td>1.53 2.00</td>
<td>1.56 1.98</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>1.51 2.03</td>
<td>1.56 1.95</td>
<td>1.57 1.91</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>1.53 2.02</td>
<td>1.51 1.92</td>
<td>1.57 2.03</td>
</tr>
</tbody>
</table>

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Table 8. ΔWRV (g H₂O/g fiber) of Whole and Fines-Free Pulps

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl.</th>
<th>CED</th>
<th>DED</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>wp</td>
<td>ff</td>
<td>wp</td>
<td>ff</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>0.47</td>
<td>0.26</td>
<td>-</td>
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<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>0.58</td>
<td>0.23</td>
<td>0.54</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>0.46</td>
<td>0.22</td>
<td>0.49</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>0.78</td>
<td>0.46</td>
<td>0.67</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>0.77</td>
<td>0.49</td>
<td>0.70</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
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<td>0.78</td>
<td>0.52</td>
<td>0.68</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>0.72</td>
<td>0.49</td>
<td>0.67</td>
</tr>
</tbody>
</table>

Table 9a. Iron Colloid μgms/gm Measurements for Fines-Free Pulps

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl. 0 revs</th>
<th>Unbl. 3000 revs</th>
<th>CED 0 revs</th>
<th>CED 3000 revs</th>
<th>DED 0 revs</th>
<th>DED 3000 revs</th>
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</thead>
<tbody>
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<td>wp</td>
<td>ff</td>
<td>wp</td>
<td>ff</td>
<td>wp</td>
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<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>12,450</td>
<td>27,000</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>16,800</td>
<td>42,100</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>21,100</td>
<td>38,600</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>18,200</td>
<td>41,100</td>
<td>22,300</td>
<td>47,000</td>
<td>22,500</td>
<td>49,200</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>20,400</td>
<td>51,100</td>
<td>22,200</td>
<td>46,100</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>21,900</td>
<td>52,600</td>
<td>22,200</td>
<td>46,100</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>20,100</td>
<td>42,500</td>
<td>22,200</td>
<td>46,100</td>
<td>22,700</td>
<td>48,800</td>
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</tbody>
</table>

Table 9b. Changes in Iron Colloid μgms/gm Pick Up with Refining

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl. ΔFe. 0 - 3000 revs</th>
<th>CED ΔFe. 0 - 3000 revs</th>
<th>DED ΔFe. 0 - 3000 revs</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0 - 3000 revs</td>
<td>0 - 3000 revs</td>
<td>0 - 3000 revs</td>
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<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>14,550</td>
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<td>-</td>
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<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>25,300</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>17,500</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>22,900</td>
<td>22,500</td>
<td>26,700</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>30,700</td>
<td>23,900</td>
<td>-</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>30,700</td>
<td>23,900</td>
<td>-</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>22,400</td>
<td>26,100</td>
<td>-</td>
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</tbody>
</table>

Confidential Information - Not for Public Disclosure
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Table 10a. Palladium Colloid μgms/gm Measurements for Fines Free Pulps

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl.</th>
<th>CED</th>
<th>DED</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0 revs</td>
<td>3000 revs</td>
<td>0 revs</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>318</td>
<td>694</td>
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<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>229</td>
<td>1180</td>
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<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>128</td>
<td>612</td>
<td>-</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
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<td>272</td>
<td>1370</td>
<td>767</td>
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<td>15.7</td>
<td>888</td>
<td>2450</td>
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<td>12.1</td>
<td>185</td>
<td>1120</td>
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<td>10.7</td>
<td>367</td>
<td>1660</td>
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</table>

Table 10b. Changes in Palladium Colloid μgms/gm Pick Up with Refining

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl. ΔPd. 0 - 3000 revs</th>
<th>CED ΔPd. 0 - 3000 revs</th>
<th>DED ΔPd. 0 - 3000 revs</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>376</td>
<td>-</td>
<td>-</td>
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<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>951</td>
<td>-</td>
<td>-</td>
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<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>484</td>
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<td>-</td>
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<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>1098</td>
<td>1543</td>
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<td>15.7</td>
<td>935</td>
<td>1562</td>
<td>-</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>1293</td>
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<td>-</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>1274</td>
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Table 11. Weighted/Weighted Average Fiber Length mm (FS-100)

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl.</th>
<th>CED</th>
<th>DED</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0 revs</td>
<td>3000 revs</td>
<td>0 revs</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>3.72</td>
<td>3.48</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>3.68</td>
<td>3.48</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>17.1</td>
<td>3.61</td>
<td>3.39</td>
<td>-</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>20.4</td>
<td>3.24</td>
<td>3.46</td>
<td>-</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>3.61</td>
<td>3.39</td>
<td>-</td>
</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>3.16</td>
<td>3.20</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>10.7</td>
<td>3.16</td>
<td>3.20</td>
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Table 12. Curl Measurements by Projection

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Unbl. 0 revs</th>
<th>3000 revs</th>
<th>CED 0 revs</th>
<th>3000 revs</th>
<th>DED 0 revs</th>
<th>3000 revs</th>
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</thead>
<tbody>
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<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>1.02</td>
<td>1.05</td>
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<td>28.1</td>
<td>1.04</td>
<td>1.07</td>
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<td>17.1</td>
<td>1.05</td>
<td>1.06</td>
<td>-</td>
<td>-</td>
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<td>-</td>
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<td>20.4</td>
<td>1.42</td>
<td>1.09</td>
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<td>-</td>
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<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>15.7</td>
<td>-</td>
<td>-</td>
<td>-</td>
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<td>-</td>
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</tr>
<tr>
<td>kappa 30 ⇒ O oxygen</td>
<td>12.1</td>
<td>-</td>
<td>-</td>
<td>-</td>
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Table 13. Unbleached Zero Span Strength Nm/g

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Wet 0 revs</th>
<th>3000 revs</th>
<th>Dry 0 revs</th>
<th>3000 revs</th>
<th>Rewet 0 revs</th>
<th>3000 revs</th>
</tr>
</thead>
<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>122.5</td>
<td>132.5</td>
<td>131.2</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>114.5</td>
<td>150.0</td>
<td>126.9</td>
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<td>-</td>
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<td>17.1</td>
<td>107.4</td>
<td>138.6</td>
<td>127.0</td>
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<td>-</td>
<td>-</td>
</tr>
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<td>20.4</td>
<td>104.1</td>
<td>144.5</td>
<td>120.1</td>
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<td>-</td>
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<td>-</td>
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<td>-</td>
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</table>

Table 14. Bleached CED Zero Span Strength Nm/g

<table>
<thead>
<tr>
<th>Material &amp; Cook</th>
<th>Unbl. Kappa no.</th>
<th>Wet 0 revs</th>
<th>3000 revs</th>
<th>Dry 0 revs</th>
<th>3000 revs</th>
<th>Rewet 0 revs</th>
<th>3000 revs</th>
</tr>
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<tbody>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>110.1</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S.P. chips ⇒ conventional kraft</td>
<td>28.1</td>
<td>104.5</td>
<td>120.6</td>
<td>122.6</td>
<td>162.2</td>
<td>109.7</td>
<td>136.1</td>
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<td>17.1</td>
<td>97.9</td>
<td>107.0</td>
<td>123.1</td>
<td>149.7</td>
<td>106.7</td>
<td>122.4</td>
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<tr>
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<td>20.4</td>
<td>105.3</td>
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<td>99.4</td>
<td>135.8</td>
<td>70.9</td>
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<td>63.1</td>
<td>81.4</td>
<td>93.4</td>
<td>134.6</td>
<td>68.4</td>
<td>92.8</td>
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<tr>
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<td>-</td>
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</tbody>
</table>
Table 15. Bleached DED Zero Span Strength Nm/g

<table>
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<th>Rewet</th>
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Table 16. Unbleached Zero Span Strength Ratios

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Table 17. Bleached CED Zero Span Strength Ratios

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Confidential Information - Not for Public Disclosure
(For IPST Member Company's Internal Use Only)
Table 18. Bleached DED Zero Span Strength Ratios

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<th>Wet/Dry 3000 revs</th>
<th>Rewet/Dry 0 revs</th>
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Figure 1. Variation of Tear Index with Kappa Number - 0 revs PFI Mill

Figure 2. Variation of Tensile Index with Kappa Number - 0 revs PFI Mill
Figure 3. Variation of Tear Index with Kappa Number - 3000 revs PFI Mill

Figure 4. Variation of Tensile Index with Kappa Number - 3000 revs PFI Mill
TABLE 19. HANDSHEET PROPERTIES - 0 REV'S PFI MILL

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<th>Basic Weight g/m²</th>
<th>Apparent Density gm/cm³</th>
<th>Gurley Porosity secs</th>
<th>Scattering Coeff.</th>
<th>Tear Index mN/m</th>
<th>Out-of-Plane Elastic Const. (M/sec²)</th>
<th>In-Plane Elastic Const. (M/sec²)</th>
<th>Elastic Index Nm/g</th>
<th>Strain Index %</th>
<th>Tensile Index Nm/g</th>
<th>Elastic Index MJ/g</th>
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**KRAFT DELIGNIFICATION**

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<th>Gurley Porosity secs</th>
<th>Scattering Coeff.</th>
<th>Tear Index mN/m</th>
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<th>In-Plane Elastic Const. (M/sec²)</th>
<th>Elastic Index Nm/g</th>
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**KRAFT AND OXYGEN DELIGNIFICATION**

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<th>Scattering Coeff.</th>
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<th>In-Plane Elastic Const. (M/sec²)</th>
<th>Elastic Index Nm/g</th>
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FUNDAMENTALS OF INTERFIBER BONDING

STATUS REPORT
FOR
PROJECT F025

Hiroki Nanko
Shaobo Pan

Institute of Paper Science and Technology
500 10th Street, N. W.
Atlanta, Georgia 30318
DUES-FUNDED PROJECT SUMMARY

Project Title: Fundamentals of Interfiber Bonding
Project Code: F025
Project Number: FBOND
PAC: Fiber and Paper Physics
Division: Fiber and Paper Physics Division
Project Staff: Hiroki Nanko, Associate Professor
               Shaobo Pan, Assistant Scientist
FY 98-99 Budget: $89,191
Time Allocation: Faculty: 38%, Staff: 50%
Supporting Research: None

RESEARCH LINE ROADMAP: Convertibility and End-Use Performance

Improve the ratio of product performance to cost for pulp and paper products by 25% by developing:
1. Models, algorithms, and functional samples of fibrous structures and coatings which describe and demonstrate improve convertibility and end-use performance, and
2. Break-through papermaking and coating processes which can produce the innovative webs with grater uniformity than that achieved by current processes.

PROJECT OBJECTIVE:
Investigate the effects of dry strength additives on structure and properties of interfiber bonding.

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PROJECT BACKGROUND:
This project started in April 1998. The purpose of this project is to understand how bonding agents work, including how bonding agents are adsorbed to the fibers, where they are located in the paper, how they contribute to bond formation, how they enhance the bond strength, and how they improve sheet strength. In order to achieve this purpose, visualization techniques of bonding agents for transmission electron microscopy (TEM) should be developed. This project will provide new insight into interfiber bonding as enhanced by the bonding agents.

SUMMARY OF RESULTS (MARCH 98- MARCH 99)
1. Visualization techniques of bonding agents were developed.
   - Colloidal gold, prepared with various recipes, was applied to the bonding agents for marking the cationic groups of the polymers.
   - Negative staining and colloidal gold techniques were combined and were optimized to visualize the bonding agent/fibril interaction on the fiber surface.
2. Bonding agents were observed.
   - Cationic starch, cationic polyacrylamide (PAM) and branched amphoteric PAM were visualized.
   - The manner of molecule adsorption of these bonding agents to the fibrils was characterized.
3. Visualization technique of the glue line structure of corrugated board was developed.

PROJECT GOALS FOR FY 1999-2000
1. Develop and improve microscopy techniques.
2. Visualization of anionic PAM.
3. Visualize natural gums.
4. Visualize the location of bonding agents in paper.
5. Examine the manner of bond breakage and evaluate bond quality.
6. Prepare reports.

PROJECT DELIVERABLES:
1. Microscopy techniques to visualize amorphous polymers.
2. Insight for design of new bonding agents.

PROJECT SCHEDULE:
April 1999 - March 2000

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INTRODUCTION
Various bonding agents are used in the paper industry today for the improvement of mechanical properties of paper. They are water soluble, hydrophilic natural, and synthetic polymers such as starch derivatives, gums, cellulose derivatives, and synthetic dry strength agents. The commercially most important are cationic starch, natural gums, and acrylamide polymers. These polymers are added to the paper stock at the wet end, and they form hydrogen bonds with pulp fibers after drying. The basic mechanism of enhancement of bond strength for such polymers is simple, "increase of hydrogen bonds". However, we do not know why polyacrylamide makes the paper stronger than cationic starch, why potato starch works better than corn starch. To answer the question, "How bonding agents work," we have to know where and how bonding agents are adsorbed on the fiber surface, where bonding agents are located in the paper, how bonding agents change the quality of interfiber bonding, and ultimately how bonding agents improve the bond strength.

On the other hand, starch has been widely used for various purposes in papermaking. It is used as a dry strength additive, as a binder for pigment coating, as an adhesive for corrugated board, and for surface sizing. In order to understand how starch works for the improvement of paper properties, it is important to know where and how the starch is located in the sheet.

BACKGROUND
The molecular conformations of the polymers directly affect the performance of polymers. Interaction between the polymers and pulp surface is also important the understanding of how polymers work. If the polymers can be seen directly by microscopic methods, we could understand the relationship between functions.
and molecular conformations of polymers better. The structure of crystallized polymers has been well investigated by X-ray Diffraction and Electron-Beam Diffraction methods. However, there are no good methods available that can visualize amorphous polymers, so we are unable to see directly the polymers to confirm whether they have expanded or coiled in solution. We can only speculate about the molecular conformation based on indirect methods such as viscosity measurement, light scattering measurement, and GPC.

Recently, Nanko and Yang [1998] successfully visualized a cationic polymer sizing agent using TEM and showed how the polymer sizing agent adsorbed onto the fibril surface (Fig.1). This polymer sizing agent is polystyrene-based

![Figure 1: Cationic polymer sizing agent adsorbed to the fibrils on the fiber surface of bleached kraft pulp. (Nanko and Yang, 1998)]
cationic copolymer. We applied a negative staining technique that is widely used to visualize viruses, cell components (e.g., ribothomes), cell fragments (e.g., membranes) and isolated macromolecules (e.g. protein). We also applied negatively charged gold colloid particles to mark the cationic sites of the polymer. Combining these two techniques, we could show clearly that the small particles adsorbed on the fibrils are the polymer sizing agent. These techniques were originally developed for transmission electron microscopy of biological materials, however, they seems applicable to the polymers used for papermaking, showing adsorption, location, and distribution in the paper.

**VISUALIZATION STRATEGY OF BONDING AGENTS**

Cationic starch consists of light atoms, such as C, O, H, and N. Such polymers cannot give enough contrast under TEM without staining by heavy metals. Electron beam also damages the polymer molecule very easily. Heavy metals can be introduced to the molecule by chemical reaction; however, the conformation of the molecules may change completely. Even if heavy metal atoms could be introduced to the molecules without changing the molecular conformation, they may not give enough density to visualize the molecule.

Use of heavy metals for visualization of polymers is inevitable for transmission electron microscopy of the amorphous polymers. In this project, however, heavy metal staining methods that never cause reaction between the stain and the polymers should be applied. Attraction forces between the stain and the polymers will be used to obtain a contrast, e.g., cationic and anionic groups of the polymers, hydrogen bonding between the stain and the polymers. The purpose of this project is not only to visualize the polymer molecules but also to know where the polymers are located and how the polymers are interacting with fibers. To achieve these purposes, the following strategies were applied.
• Used TEM as a main tool to observe the interaction between fibers and the polymers at the fibril level.
• Apply negative stain technique to visualize the fibrils and the polymers adsorbed to the fibrils.
• Use colloidal gold to detect cationic groups in the polymer.

In practice, the above-mentioned techniques should be modified to meet the specific purposes of the research and specific properties of the polymers.

METHODS FOR VISUALIZING BONDING AGENTS

1. Pulp preparation

A dry lap southern pine bleached kraft pulp was used. The pulp was beaten by PFI mill to 500ml CSF. After beating the pulp was washed by Nanopure water several times and used for sample preparation.

2. Application of cationic starch

Cationic starch made of corn starch was used. Cationic starch was added to the pulp slurry at the levels of 0.5% and 1.0% based on dry pulp and dry starch. The slurry was stirred for 10 min after the cationic starch solution was added.

3. Application of PAM

• C-PAM:
  - Molecular weight: 1,000,000
  - Ion density: 0.47 meq/g
  - Cationic group: \(-\text{CONH}(\text{CH}_2)_3\text{N}^+\langle\text{CH}_2\rangle_3\text{H}...1/2(\text{SO}_4)^2^-\)

• Amphoteric PAM (branched):
  - Molecular weight: 1,800,000
  - Ion density: anion: 0.64 meq/g, cation: 0.77 meq /g
  - Cationic group: \(-\text{CONH}(\text{CH}_2)_3\text{N}^+\langle\text{CH}_2\rangle_3\text{H}...1/2(\text{SO}_4)^2^-\)
Anionic group: -COOH

PAM was added to the pulp slurry at the level of 0.3% based on dry pulp and dry polymer solids. The slurry was stirred for 10 min after the PAM solution was added.

4. Preparation of gold colloid

Monodispersed gold colloid particles of the 3 – 4 nm diameter were prepared reducing chloroauric acid by trisodium citrate.

5. Marking of cationic sites of polymer by gold colloid

Colloidal gold was added to the pulp/C-PAM suspension or to the pulp/C-starch suspension. After stirring for 1 min, the suspension was left to stand for 10 minutes before negative staining.

6. Negative staining

General principles of negative staining are as follows:

- Prepare grids with carbon-coated supporting film.
- Make the supporting film surface hydrophilic by ion etching.
- Place a droplet of pulp suspension on a grid and remove water using filter paper.
- Drop 4% uranyl acetate solution on the grid, remove the solution by filter paper, and air dry the sample.

The heavy metal remains surrounding particles on the supporting film so that the shape of the particles can be seen in the dark background under the TEM. There is no reaction between the stain and the sample. Uranyl acetate and phosphotungstic acid were used as negative stains in this project.
ADSORPTION OF C-PAM AND C-STARCH TO FIBRILS

Figure 2 is a control sample showing negatively stained fibrils generated by beating of softwood bleached kraft pulp. The fibril surface looks very clean and without any contamination. Individual cellulose microfibrils can be seen clearly by this technique. In this research, the interaction between fibers and the bonding agents was examined at fibril level because the refined fiber surface is covered by the fibrils and the fibrils are the actual adsorption sites of the bonding agents. It is also known that a large proportion of the bonding agent is adsorbed to secondary fines. Secondary fines are detached fibrils from the fiber surface, having the same morphology as the fibrils on the fiber surface.

Fig. 2. Negatively stained secondary fines. Scale: 0.1 μm.
1. Cationic Starch

Figures 3 and 4 are the negatively stained fibrils with adsorbed cationic starch. Gold colloid particles of 3 - 4 nm are showing the location of the cationic starch. When cationic starch was added by 0.5%, the gold particles partially covered the fibril surface (Fig. 3). The gold particles were also found spreading into the vicinity of the fibrils from the adsorbed sites. This suggests that the cationic starch molecules adsorbed on the fibrils in the loosely agglomerated state. The fibril surface is just partially covered by the cationic starch. When cationic starch was added by 1.0%, most of the fibril surface was covered by agglomerated starch (Fig. 4). The agglomerated starch looked dense and tightly adhered to the fibril surface.

Fig. 3. Cationic starch adsorbed to the fibrils. Scale: 0.1 \( \mu \) m.
Fig. 4. Cationic starch adsorbed to the fibrils. Scale: 0.1 μm.

2. Cationic PAM

Cationic PAM was added to the beaten pulp by 0.3% and stained by the colloidal gold. Figures 5 and 6 are the negatively stained image of the adsorbed C-PAM to the fibrils. Unexpectedly we found many straight dark lines extend from the fibril surface toward the outside. These lines were 0.5 to 1.5 μm long. They are almost straight and parallel to each other. Gold particles are sparsely aligned on the lines. Some lines are thicker than the other. Gold particles are densely aligned on the thicker lines. The root of the lines where they attached to the fibrils are thicker and they tend to be thinner forming branches toward the outside. These lines should be stretched cationic PAM molecules. Thick lines should be bundles of C-PAM molecules and thinner lines should be thinner bundles of C-PAM molecules. The thinnest one could be a single C-PAM
Fig. 5. C-PAM adsorbed to the fibril surface. Scale: 0.1 μ m.

Fig. 6. C-PAM adsorbed to the fibril surface. Scale: 0.1 μ m.
molecule. As shown in Fig. 7, C-PAM molecules form a network between fibrils. Gold particles were also found on the fibrils, however, they did not deposit in the agglomerated state on the fibril surface. It appears that a part of the C-PAM molecules attached to the fibril surface at one end and the main body of the molecules is stretched outwardly away from the fibril.

![Fig. 7 C-PAM molecules forming network between fibrils. Scale: 0.1 μm.](image)

We observed the conformation of individual molecules. As shown in Fig. 8, the C-PAM molecules were almost straight. There were lines between gold particles in some cases, but such lines could not be seen in most of the cases. The length of the molecules, however, is still measurable judging from the aliened gold particles. We measured the length of some molecules on the TEM micrographs. For example, the molecule shown in Fig. 8 is 1.47 μm long. The longest molecule we measured was 1.9 μm long. The molecular weight of

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the C-PAM used for this experiment is 1,000,000. The average length of the molecule calculated from the molecular weight of the C-PAM is 3.541 μ m. This value is a little longer but in the same order as the measured length of the molecule.

In the case of the molecules in Fig. 8, the distance between gold particles varies from 125 to 5 nm. The average distance between two cationic groups calculated from the charge density of the C-PAM is 7.2 nm. This means that the marked cationic groups by the gold particles are much less than the real number of cationic groups.

Fig. 8. C-PAM molecules Scale: 0.1 μ m.
The C-PAM molecules in the micrographs are in the extended form. We have to know whether the C-PAM has such conformation in the aqueous solution or not. There is a possibility of artifact that the gold particles adsorbed to the fibrils distort the conformation of C-PAM. Strong repulsive force between negatively charged gold particles could make the molecules straight. However, this may not be the case because the dark lines without gold particles are also in the extended form. The repulsive force that works between cationic groups on the C-PAM molecules may be enough to keep the molecules straight.

Another mystery of the image is that all the lines are aligned in one direction. The molecules might be aligned by the movement of water when the water on the grid was sucked by the blotter paper in the sample preparation process. It can be assumed that the C-PAM molecules are extended radially from the fibrils in the solution. Such molecules can be aligned and straightened when the water is sucked in a certain direction.

It was expected that the molecules should be well separated from each other because of the repulsive force that works between molecules. However, the thick dark lines suggest that the C-PAM molecules form bundles. The branched structure of dark lines in Figs. 5 and 6 may suggest the longitudinal entanglement of the linear molecules. The C-PAM molecules look long enough to get entangled easily in the solution. It is not clear whether the C-PAM molecules form bundles in the aqueous solution or such bundles are formed in the drying process. We suspect that the longitudinally entangled molecules get closer and form bundles each other due to water surface tension force during drying. Because of the low charge density of the C-PAM, the repulsive force may not be strong enough to prevent such bundle formation.
It was not expected that the C-PAM can be positively stained by uranyl acetate. The C-PAM can be stained only under specific conditions. Now, we are working on improving the staining method to achieve a stable and constant staining effect. We do not understand the staining mechanism yet, however, this method will give us a clue to understand the molecular conformation of the water soluble amorphous polymers.

3. Amphoteric PAM (branched)

Branched amphoteric PAM was added to the beaten pulp by 0.3% and stained by the colloidal gold. Amphoteric PAM adsorbed to the fibrils differently from the C-PAM. Thin dark lines were observed close to the fibrils (Fig. 9), however, they were much shorter than the C-PAM (Figs. 5 and 6). They were only 0.2 to 0.4 μm long. This amphoteric PAM has a branched structure. Branches of the PAM molecules may have prevented the formation of thick and long bundles.
Fig. 9. Branched amphoteric PAM adsorbed to the fibril surface.

Scale: 0.1 μm.

of the polymers. Many gold particles were found widely distributed in the outside of the short lines. This may suggest the PAM molecules that are directly adsorbed to the fibrils are capturing free PAM molecules electrostatically on their outer boundary.

CONCLUSIONS
We developed a technique that can visualize bonding agents under TEM combining the negative staining method and the colloidal gold marking method. This technique showed that cationic starch and C-PAM are adsorbed to the fibrils in different manners (Figs. 4 and 5). We haven't completely understood what these images are telling us yet. We also haven't eliminated the possibility of artifacts contained in these images yet. However, it is obvious that this technique provides new insight into the conformation of polymers in the aqueous solution as well as to the polymer/fibril interaction.

The molecules of cationic starch agglomerate and deposit on the fibril surface. On the other hand, the molecules of C-PAM anchor to the fibril surface at one end, but the other end is extended out toward the solution. If these are not artifacts, we expect that these two bonding agents have completely different mechanism of bond strength enhancement. Cationic starch can enhance bonding only on the fibril surface by contact. However, a long molecule of C-PAM can bond to as many fibers and fines as it can reach. As a result, all fines and fibers become connected in the sheet by the network of C-PAM molecules. Such a difference of quality of bond enhancement mechanisms should be the reason C-PAM can make the tensile strength of paper stronger than cationic starch. Extended conformation of C-PAM molecules can form hydrogen
bonding more efficiently than the agglomerated starch molecules. This is one of the reasons C-PAM works better than cationic starch at much lower dosages.

**FUTURE WORK**

We will keep working on the C-PAM and cationic starch to establish the visualization techniques. New techniques should be developed for the natural gums and anionic PAM. We are also planning to visualize the location of bonding agents in the paper to show how bonding agents enhance the bonding. By examining by the difference in bond breakage pattern, we will better understand the quality of the bonds.