

ABSTRACT

Converting Coconut Husks into Binderless Particle Board

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Coconuts grow abundantly in tropical regions, and the coconut husk is often discarded as waste. However, a new application has increased interest in developing this previously underutilized resource. Coconut pith has chemical reactivity due to its high lignin and phenolic content and can be hot-pressed into binderless particle board with excellent commercial potential. This initial investigation into proof of concept and production details determines the key processing variables and discusses the important findings of the concurrent research done by the Common Fund for Commodities. The objective of this research is to identify the best production paths to make high quality "Cocoboard" at the lowest possible price.

Converting Coconut Husks into Binderless Particle Board

by

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A Thesis

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DEDICATION

To my wonderful wife, children, family and Jesus, our Lord, who makes all things possible.

CHAPTER ONE

Introduction

1.1 Opportunities

Within 1,400 miles of the equator there exist an abundance of both poverty and unused coconuts. This region is underdeveloped and home to many of the world's poorest populations. In this same region grow 50 billion coconuts each year, 95% of which are owned by farmers that make less than \$2 per day. It is estimated that 11 million farmers rely exclusively on coconuts for their income. Including children and extended family members, the number of people surviving on meager coconut sales may be as high as 80 million. If coconuts could be developed, both technically and commercially, into high value applications, the price of the coconut commodity would rise and increase the income and quality of life for millions around the globe [1].

1.2 Coconut Uses

When most people think of the coconut fruit, they picture only the mature hard brown nut that is typically sold in grocery stores. This is only part of what is available to the people of the tropics. The constituent parts of the coconut each have their own unique properties that could make them commercially attractive. Figure 1.1 summarizes the important properties of each part of the coconut.

On the inside of a mature nut is the edible white meat called copra that is an excellent source of nutrition. Its short chain saturated fat is an excellent source of healthy lauric acid and medium-chain triglycerides. Coconut oil is a very efficient fuel source for the body and lauric acid, which give anti-viral and anti-bacterial properties, is the main constituent [2]. Coconut oil is widely produced and sold

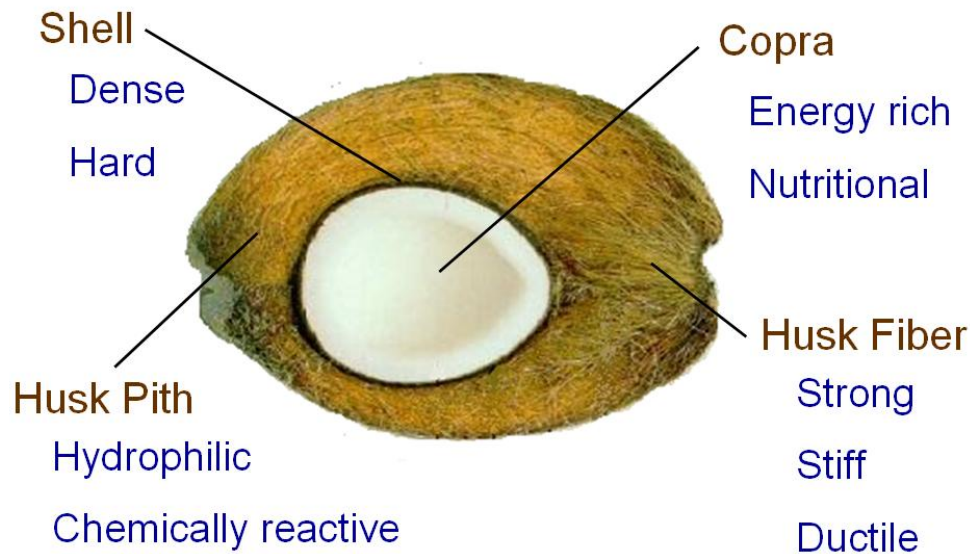


Figure 1.1: Parts of the coconut and their unique properties

throughout the tropics as food, as ingredients for hair and skin care products and as fuel for modified engines.

The coconut shell has a density of about 1.2 g/cm^3 and is five times harder than the hardest hardwood grown in the United States. The extremely high density and hardness make coconut shell an excellent feed stock for charcoal and activated carbon filters. While the shell is widely used for these applications, the price of charcoal is low and demand for activated carbon filters utilize only a small fraction of the coconut shell that is available.

The outer husk that is removed before export is comprised of long, rough fibers held together by a dust like pith as seen in Figure 1.2. The fibers are made into a variety of products such as floor mats, roofing, mattress stuffing and geotechnical netting used for erosion prevention. The fiber has an excellent combination of strength, stiffness and ductility that make it an excellent candidate to replace oil based synthetic fibers in polymeric composites. This new applications is just starting to be introduced into U.S. industries and holds the potential to greatly increase demand for coconut fiber.



Figure 1.2: Piece of coconut husk with its constituent parts

The pith is extracted from the husk along with the fiber. It has two interesting properties that make it commercially attractive. Pith is both highly hydrophilic and chemically reactive. A micrograph of coconut pith can be seen in Figure 1.3 that shows its internal structure of thin hollow shells. These hollow tubes allow for the pith to absorb ten times its own weight in water and make it an excellent additive for gardening soil. There are some synthetic materials that can absorb more water, but they are expensive and are not bio-degradable.

The pith's other property, the chemical reactivity, is the subject of this study. Naturally occurring chemicals in the pith allow it to be hot pressed into a binderless particle board that may be used as a wood substitute in places where wood resources are scarce and costly or where there are market demands for green materials. It has been said that man has made everything from pith except money, but that notion is going to change. Coconut pith will soon be produced into a viable building product that will help raise the value of coconuts and improve the income of millions of coconut farmers around the world.

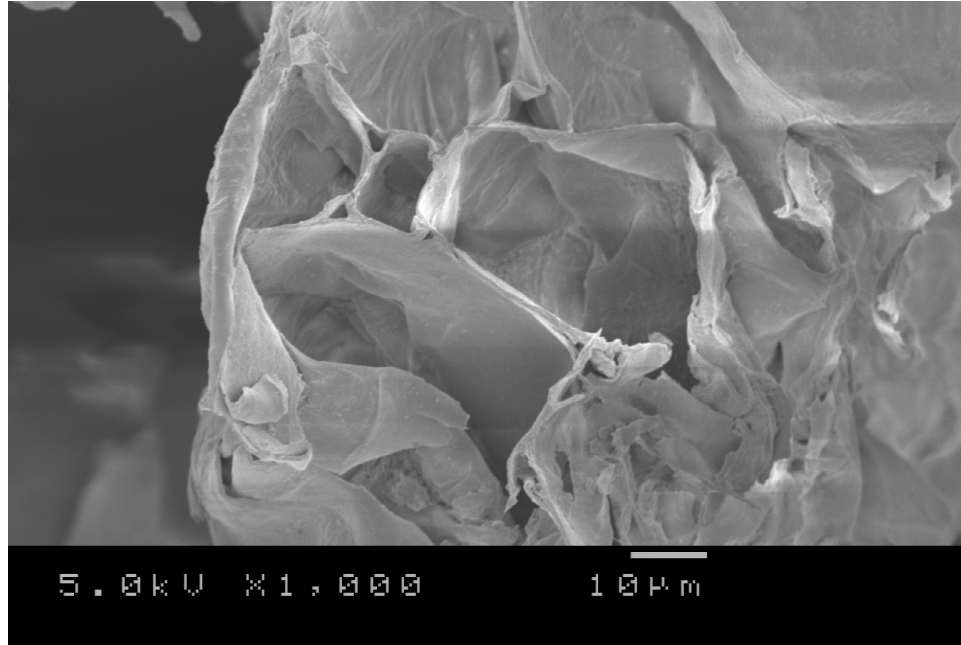


Figure 1.3: Micrograph of coconut pith particle showing internal structure.

1.3 Wood

Wood is an abundant natural resource with unique characteristics that have made it a choice building material throughout history. It has been made into weapons, tools, shelter, transportation, art and many other uses. Today, wood is still valued for its beauty, workability and versatility.

The properties important in selecting a wood include density, appearance and mechanical properties. The properties of different species change with the relative amounts of cellulose, lignin, hemicelluloses and other basic compounds contained within their cellular structures. While wood properties remain mostly constant, enough variations can exist within trees of a single species and within individual trees that engineered wood, made by purposefully joining small wood pieces together, is finding a growing place in wood markets.

The properties of engineered wood can be varied over a wider range and with better consistency than natural wood. The best wood in nature can, for some appli-

cations, be beat with carefully controlled production processes since the properties are determined on a mesoscopic level. That is, instead of being determined by the contents of the cell, engineered wood properties are determined by the properties, organization and combination of fibers, veneers, chips or particles.

Particleboard is a composite material that is describe as any woody material that is adhesive-bonded together. It is among the fastest growing wood-based industries, along with other engineered woods like plywood, oriented strandboard, hardboard, and cellulosic fiberboard. Particle board, like other composite woods, is used in structural applications such as sub-flooring, furniture and support structures and nonstructural applications such as interior covering panels.

The process to make wood composites usually consists of gluing fiber pieces together with a binder, forming them into a mat, and pressing them into the final product. The binder is typically a thermosetting or heat-curing resin often made from toxic formaldehydes. These toxins will continue to be used unless the supply of petrochemicals is interrupted or a new adhesive system is developed from renewable resources [3].

1.4 Research Background

It was understood, at least as early as 1978 in Indonesia [4] that coconut coir has potential for use in structural components. For the next twenty years, very little was done outside of India to realize the value of this agricultural waste product until the late 1990's. In July 1997, Dr. Jan E.G. van Dam presented a paper titled "Prospects for coir technology and market developments" at a coconut technology meeting in the Philippines. Not long after this Dr. van Dam was funded to further investigate coir building and packaging material in the form of binderless particle board. He produced a series of three papers on the subject and subsequently a final technical report, which we obtained in a by-invitation-only meeting in the

Philippines, midway through this research program, in October 2007. It allowed us to compare our result, and it also raised additional interesting questions which we explored.

In 2004, Dr. John Pumwa spent a sabbatical at Baylor University researching the electrification of rural villages with coconut bio-diesel with Dr. Walter Bradley. During their research they quickly realized that it takes 37 coconuts to make a gallon of coco-biodiesel, producing 37 husks and shells that are either a waste disposal problem or free feedstock for other yet-to-be discovered products. With a goal of helping rural villages find more value from every part of the coconut, they began research on existing and new methods for processing coconuts and business plans, with the assistance of Dr. Jeffrey McMullen in Management and Entrepreneurship at Baylor, to increase economic opportunity in rural villages. After Pumwa returned to Papua New Guinea, Bradley continued the research and planning for commercialization with Baylor engineering and business students. Research on producing binderless particle board from coconut husks started at Baylor in the spring of 2006.



Figure 1.4: Typical pile of unused coconut husks in coconut producing regions.

1.5 Process Overview

Both coconut fiber and pith have lignin with extremely high phenolic content [4]. When the pith is heated under pressure to 130°C the lignin melts and cross-links like a thermosetting phenolic resin. This allows for the production of three-dimensional molded parts or more importantly, organic binderless particle board [4]. Because coconut pith is a natural agricultural product, its behavior during processing is complicated. At different points in processing the pith shows different combinations of traits of unconsolidated agricultural mass, thermosetting resins and consolidated agricultural mass. Thus it is essential to identify the most important processing variables. In order for Cocoboard producers to succeed, a complete understanding of how processing controls the properties of the final product is required [3].

1.6 Project Scope

The scope of this project is to gain an initial understanding of the processing of coconut pith into binderless particleboard and the role processing variables play in determining the properties of the final product. This project sorts production factors of varying levels of importance based on an analysis of previous work and new experiments. The progression of experiments performed for this initial investigation is presented along with selected results of test performed at Baylor University. Additionally, results from van Dam's research will be included where appropriate.

CHAPTER TWO

Literature Review

2.1 R. Viswanathan and L. Gothandapani

The earliest available papers on the subject of processing coconut pith were produced from Tamil Nadu Agricultural University in India in the late 1990's. In a series of four papers, Viswanathan reports a natural progression of experiments to determine the effectiveness of making coir boards using two different resins as glue. The first paper discusses the relationships between pressure and density as well as stress relaxation characteristics of coconut pith, but since the mechanism of solidification using a resin is fundamentally different from using the coir's own natural binder, the results of Viswanathan's other papers are not applicable to this project.

Viswanathan starts with an analysis of the mechanical relaxation properties of coconut pith. He says, "Compressing the biomass to densify into any form for the production of briquettes or particle boards requires an understanding of the behaviour of the material under pressure in the die and after it has been removed from the load. Hence, the pressure density characteristics and stress relaxation behaviour of coir pith are necessary for calculating the force required to compress the pith to the desired density and for the design of the compressing system [5]."

While this is true and would normally be useful, the methods and analysis do not account for an important factor. It is believed that the authors were unaware of the pith's natural thermosetting behavior because there was no mention of the heat induced viscosity changes or self-adhesion property.

2.2 *Jan E.G. van Dam, et al.*

The only research reported in the literature on pith building and packaging material in the form of binderless particle board was sponsored by the Common Fund for Commodities: Coir Based Building and Packaging Materials, (CFC/FAO-IGHF/11) and conducted by Jan E.G. van Dam, et al. The results were reported in a series of papers on the subject and subsequently a final project report which we obtained in 2007.

Van Dam's proof of concept paper was an outlook on the potential of developing a sustainable building material as a timber substitute. They concluded that this valid concept showed economic feasibility, environmental benefits and potential for socio-economic development in coconut producing regions [6]. The other papers detailed the development of a process for production of high density, high performance binderless boards from whole coconut husk.

In part one, van Dam investigated the properties of lignin as a thermosetting binder with infrared spectroscopy and scanning electron microscopy. This initial investigation of the morphology and chemical composition of coconut husk, fiber and pith found that the non-crystalline change in viscosity and the potential for cross linking at elevated temperatures are key issues if the pith is to be used to make binderless particleboard. The exploration of the chemical reactivity of the pith concluded that the husk material undergoes an irreversible softening that indicates the thermosetting behavior of the lignin and under controlled thermal conditions may be utilized to develop a process for production of binderless boards [4].

In part two, van Dam further investigated the coconut husk morphology, composition and properties. Husk anatomy, density, tensile properties, swelling, and chemical composition of the coconut husk components were analyzed, and he found that differences in types of coconuts were negligible but that age may have some impact on the processability of the husks [7].

These and additional results were explained in greater detail in the final report for project CFC/FIGHF/11 [8]. The report included details on the effect of raw materials and processing on product properties, the up scaling of the production process, pilot trial results, and an evaluation on the economic and marketing aspects. A detailed discussion of the applicable results of this technical report is presented in Appendix A.

CHAPTER THREE

Experimental Materials and Methods

3.1 Materials and Equipment

Pictures of the equipment have been compiled and can be found in Figures 3.1 through 3.14.

3.1.1 MTS Electro-Mechanical Testing Equipment

An MTS Q-Test 100 frame controlled by TestWorks software was used for most hot pressing experiments. An oven with a fan and electric heaters was mounted on the MTS frame. A 100kN load cell measured the loads that were transmitted by a steel cylindrical rod extending from the cross head into a hole in the top of the oven. Different dies were machined to be mounted inside the oven on the bottom attachment. Electricity was provided to the heaters from a standard wall plug through a Dawyer Series 16A Temperature Controller. Temperatures were recorded using LabVIEW software on a separate computer. Various load and temperature paths were obtained by controlling the heaters and MTS crosshead. Mechanical property tests were performed on the MTS frame as well. For these tests, a 10kN load cell measured the loads applied to the 3.5" span three-point bending test apparatus.

3.1.2 Dake Lab Press

Boards, 6" x 9", were produced on a 25 ton Dake lab scale press (model number: 44226) with a manually actuated cylinder and dial controlled hot platens. A cut baking sheet and a rectangular frame were used to evenly distribute pith on the bottom platen.



Figure 3.1: Q-Test 100 with punch and die.

3.1.3 Scanning Electron Microscope

A JEOL JSM-5410 scanning electron microscope was used to create micrographs. A Denton Vacuum Desk II sputter coater was used to coat specimens after they were dried in a Precision gravity convection oven.



Figure 3.2: Assembled view of oven and heating elements.

3.2 Methods and Procedures

3.2.1 Pith Acquisition

Three different types of pith were studied. Pith from husks from coconut trees in Mexico was hand separated from the husk fiber with a pet brush. Pith from the same Mexican coconut trees was machine separated and ordered in bulk. Alternatively, coconut pith from The Philippines pressed into bricks was purchased from a garden supply company. The compressed bricks were broken into particles by hand, soaking in water and drying or by grating with a handheld cheese grater.

3.2.2 Hot Pressing

For each experiment performed on the MTS equipment, pith was massed and hand pressed between two cross section pieces inside a die. Thermocouples were placed through holes in each end and the die and connected to the temperature controller and the LabVIEW hardware. The die was placed on the bottom mounting bracket, and the heaters were place in each corner before the door was secured with a threaded nut and bolt. The fan and temperature controller were plugged into standard wall plugs.



Figure 3.3: Die and load transfer pieces.

The load path was controlled with different versions of TestWorks software. The temperature path was controlled by plugging the heaters into a grounded standard wall plug through the temperature controller. Load and temperature data were recorded with the TestWorks and LabVIEW software respectively.

Once the desired processing conditions were met, the heaters were unplugged, the front heaters were removed through the opened oven door and the fan was left on. The die was able to be removed with heat resistant gloves once the mounting bracket and die cooled enough to remove the thermal expansion interference. Two metal punches were used to push the sample from the die through the bottom holes and the thermocouples were removed. The sample was massed, labeled for identification on both ends and left to cool to room temperature.

The load and size restrictions of processing on the MTS equipment allowed only 1" x 5" samples to be made. For this reason, the Duke lab press was used to make larger 6" x 9" boards. To do this the rectangular frame was placed on the baking sheet and filled with pith. The loaded baking sheet was placed on the bottom platen, and the frame was removed so that the pith could relax naturally on

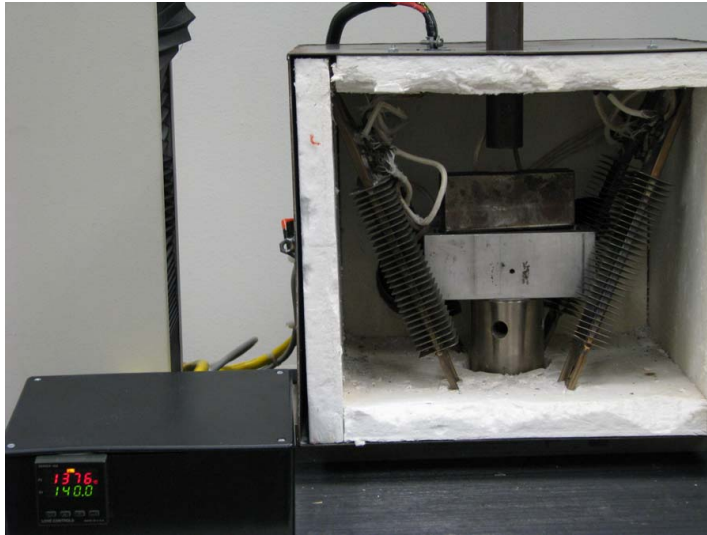


Figure 3.4: Complete experimental assembly.

the baking sheet. Various loads were applied with the manually activated hydraulic system and preheated platens were used to achieve various temperatures.

Once the desired processing conditions were met, the sample was removed from the load and heat by placing it to cool untouched on a flat surface. Once at room temperature, the uncompressed edges were removed with a band saw. Appropriate bend specimens were cut from the sample and tested in the same manner as the MTS produced samples.

3.2.3 Mechanical Properties Characterization

All samples, either from the 5" x 1" die or cut from the 6" x 9" boards, were subjected to a three-point bend test in accordance with ASTM D7031. Flexure modulus and strength were calculated using standard relationships from ASTM D7031 from the measured load-displacement test data.

Additionally, Brinell hardness tests were performed on each sample. Because of the soft nature of the samples, a Rockwell Hardness Tester was used to place only a 15 kg load on each sample with a 0.25" or 0.125" ball. The diameters of the indentions were measured with a dial caliper viewed under a binocular microscope.

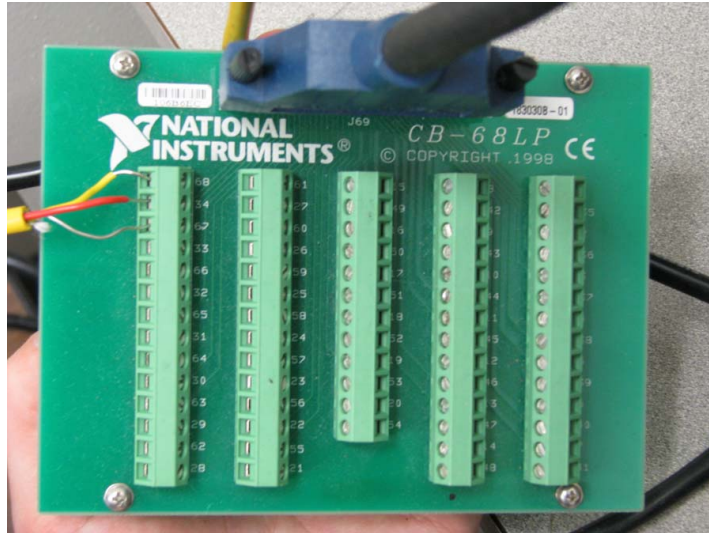


Figure 3.5: LabVIEW hardware setup.

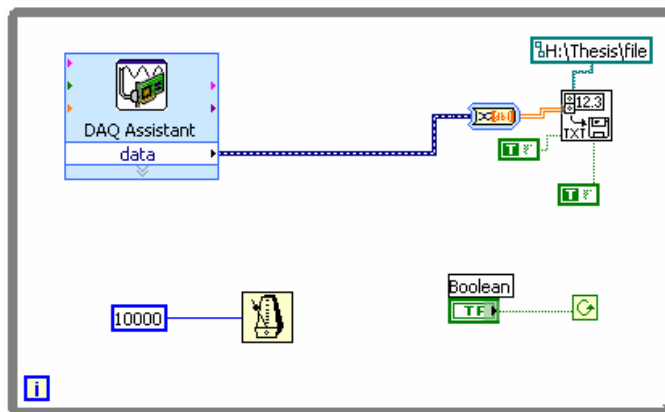


Figure 3.6: LabVIEW block diagram.

The hardness values were calculated as kilograms per square millimeter with the standard equations.

3.2.4 Characterization of Pith Morphology

Pith samples from all three sources were dried in the lab oven at 70°C and mounted on small aluminum stubs with double stick tape grounded with silver paint. The stubs were sputter coated with gold and the microstructures were viewed in the SEM lab of the Baylor science building.



Figure 3.7: Dake lab press.



Figure 3.8: Pith frame.



Figure 3.9: Precision gravity convection oven.



Figure 3.10: Sputter coater.



Figure 3.11: Scanning electron microscope.



Figure 3.12: Rockwell hardness tester.

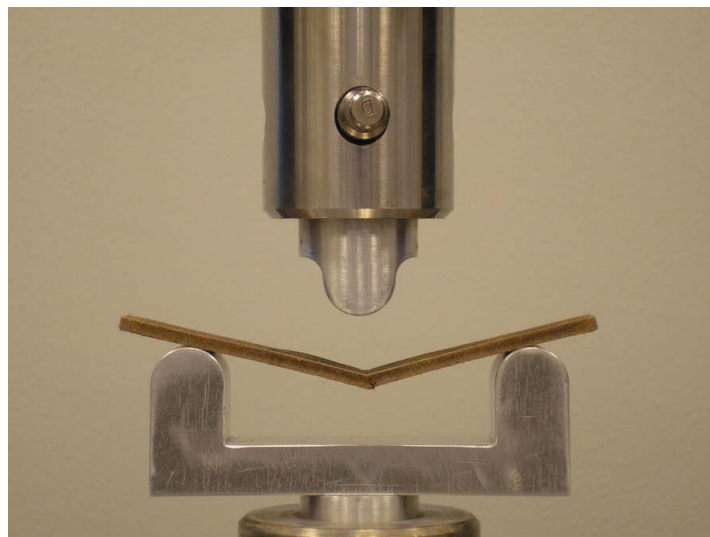


Figure 3.13: Three-point bend testing apparatus.

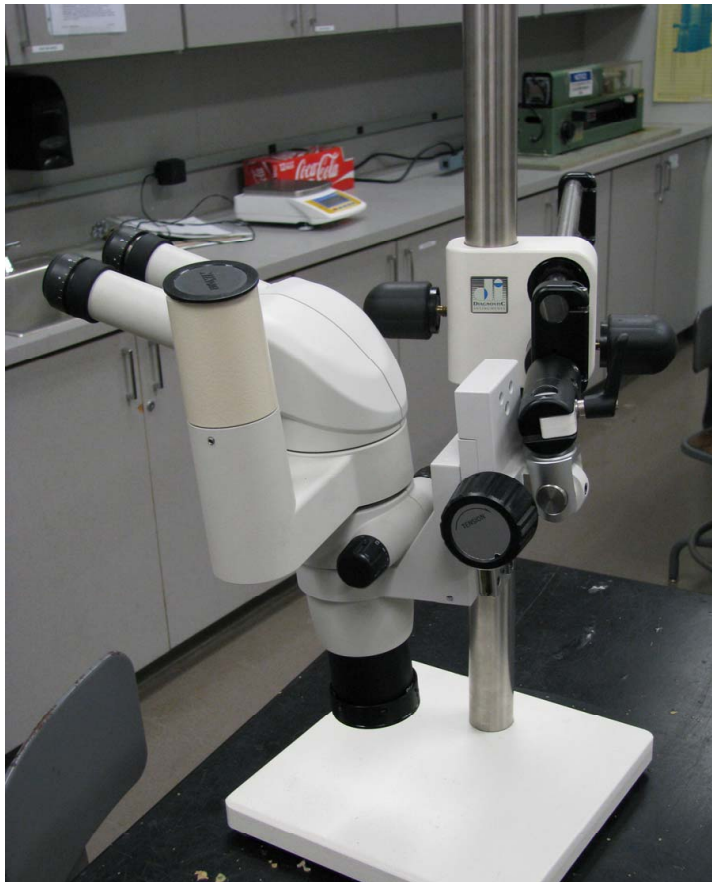


Figure 3.14: Binocular microscope.

CHAPTER FOUR

Experimental Results and Discussion

4.1 Proof of Concept

The initial papers published by van Dam clearly suggested that pith hot presses like a thermosetting resin, which would give good promise to make binderless particle board. Our initial efforts in this work were to confirm the thermosetting behavior of the pith and the possibility of making binderless particleboard.

4.1.1 Testing of Thermosetting Properties

A differential scanning calorimeter (DSC) scan of coconut husk from van Dam is shown in Figure 4.1 [4]. It indicates an exotherm around 140°C that implies a thermosetting chemical reaction with cross-linking. Hand separated pith from mature coconut husks from Mexico was tested three times with a DSC at Texas A & M University in an attempt to confirm its thermosetting properties. None of the three scans scan showed any exothermic or endothermic reaction.

Due to the lack of chemical reaction, it was assumed that the more recent scans were correct and that cross-linking does not occur. This led to the conclusion that the coconut pith behaves as a thermoplastic instead of a thermoset as indicated in the literature.

4.1.2 Round Disk Samples

Because of the assumption that the pith behaves as a thermoplastic, different combinations of final temperature and pressure were tested to find the optimal processing conditions. The initial proof of concept experiments were performed with hand separated coconut pith pressed in a one inch diameter steel die. Seventeen were hot pressed at various combinations of temperature and pressure with assis-

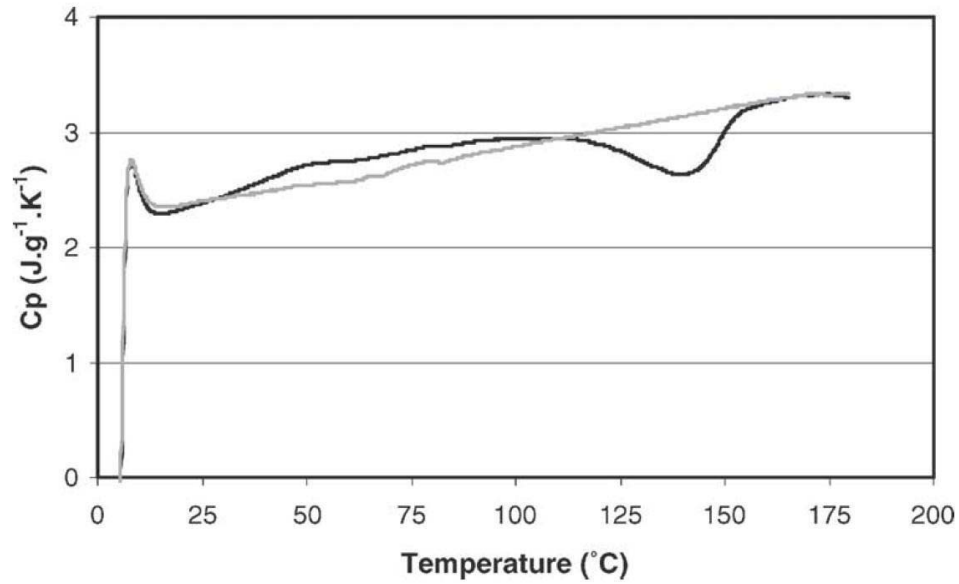


Figure 4.1: Cp - thermogram of coconut husk. First and second heating ramp [4].

tance from Jason Poel and Gideon Jeffrey. The data is summarized in Table 4.1. The relationship between density and hardness was graphed in Figure 4.2.

General trends were seen between the processing methods and the resultant densities. The mechanical properties of hardwoods are generally considered to be monotonic to density, and the same increase in hardness with density was observed for the disk samples [3].

The initial experiments, performed to gain a general understanding of the pith's behavior during processing, applied the pressure before the temperature. Specimens 2-9, 14, and 17-28 were processed by this method and create the middle range of densities.

Due to the thermoplastic assumption, we thought the processing paths were independent so that only the final combination of temperature and pressure were important. However, for samples 12, 13 and 16, the heat was applied before the pressure and this method resulted in a lower density range than the initial method. Only an upper limit could be taken on the hardness because of the extreme softness.

Table 4.1: Data from successful coconut disk specimens.

Test #	Processing Conditions			ρ g/cm ³	Brinell
	F lb	T °C	t at max min.		Hardness kg/mm ²
2	150	150	10	0.528	1.01
3	200	150	10	0.582	0.84
4	300	150	10	0.662	1.38
5	300	150	10	0.753	2.15
6	300	160	10	0.684	1.93
9	400	160	20	0.775	2.28
12	400	160	10	0.434	<0.24
13	300	160	20	0.482	<0.24
14	300	160	20	0.661	1.37
16	300	180	45	0.463	<0.24
17	300	180	10	0.698	1.37
18	300	200	10	0.701	1.55
22	400	160	35	0.75	1.37
23	400	200	-	0.766	2.35
26	400	160	-	0.909	2.32
27	400	160	-	0.921	2.32
28	400	160	-	0.88	2.32

Samples 26-28 were made with the pressure applied first, but from water saturated pith. These created the higher range of densities, which would be expected if water acts as a plasticizer that allows for better mechanical interlocking before being driven away by the heat [9]. The data from Table 4.1 was sorted according to density and tabulated in Table 4.2.

For these experiments, processing order, discussed in the previous section, and initial moisture content seemed to have an impact on properties along with processing temperature and pressure combinations as there was no overlap in the density ranges seen in Table 4.2.

4.1.3 Three-Point Bend Samples

Once we had shown that the concept of making solid wood pieces from coconut pith was valid, we needed samples appropriate for mechanical testing to gain a better understanding of the relationship of processing to final properties.

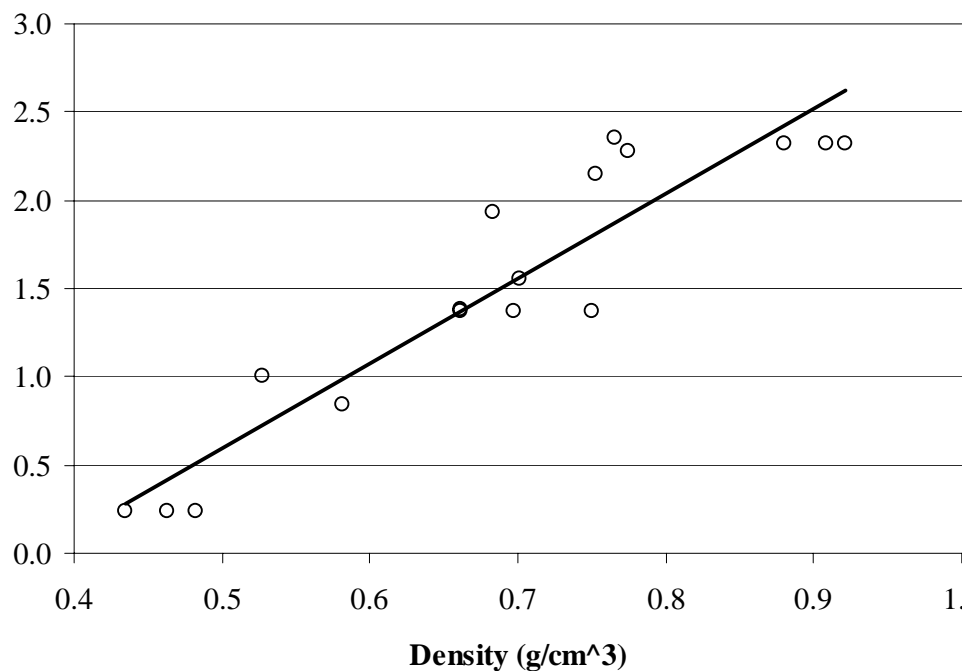


Figure 4.2: Comparison of Brinell hardness and density of coconut board disks.

Initially, specimens suitable for ASTM bend testing were made with hand separated coconut pith with the rectangular 5" x 1" aluminum die. Tests 1 - 8 were performed under load control where the pressure was held constant and tests 9 and 10 were displacement control test where the pith was compressed and held at a constant thickness. Displacement control tests provide a good understanding of the pith viscosity changes during heating because there is a fixed amount of room for the pith to expand and contract. For the displacement controlled test graphed in Figure 4.3, the male half of the die was used to press the pith in the die to a preset displacement which produced a load of 3000 lbs of compression at ambient temperature. Next the die and the pith inside were heated to a preset temperature. In displacement control tests, if the pith thermally expands, the compressive load rises since there is no room for expansion. This can be understood in the same way as the pith thermally expanding and then being recompressed back to its original thickness with an incremental increase in compressive loading. Conversely, if the

Table 4.2: Ranking of Table 4.1 data according to density.

Test #	Processing Conditions			ρ g/cm ³	Brinell	Method
	F lb	T °C	t at max min.		Hardness kg/mm ²	
12	400	160	10	0.434	<0.24	2
16	300	180	45	0.463	<0.24	2
13	300	160	20	0.482	<0.24	2
2	150	150	10	0.528	1.01	1
3	200	150	10	0.582	0.84	1
14	300	160	20	0.661	1.37	1
4	300	150	10	0.662	1.38	1
6	300	160	10	0.684	1.93	1
17	300	180	10	0.698	1.37	1
18	300	200	10	0.701	1.55	1
22	400	160	35	0.75	1.37	1
5	300	150	10	0.753	2.15	1
23	400	200	-	0.766	2.35	1
9	400	160	20	0.775	2.28	1
28	400	160	-	0.88	2.32	3
26	400	160	-	0.909	2.32	3
27	400	160	-	0.921	2.32	3

pith relaxes or contracts, the compressive load will decrease with the loss of pressure exerted on the die from the pith.

Eight specimens were suitable for comparison. Their mechanical properties are summarized in Table 4.3, flexural modulus and strength have been graphed against density in Figure 4.2, and Brinell hardness data for all specimens made from hand separated pith have been graphed against density in Figure 4.5.

Table 4.3 and Figures 4.4 to 4.5 show positive associations between density and all three measured mechanical properties, which follow the rule of thumb for hardwoods. Based on this trend and the thermoplastic assumption, we decided that future processing should be focused on controlling density and maximizing properties for a given density.

We observed thermosetting tendencies during the proof of concept tests. The observed thermosetting behaviors included path dependant processing, unrepeatable

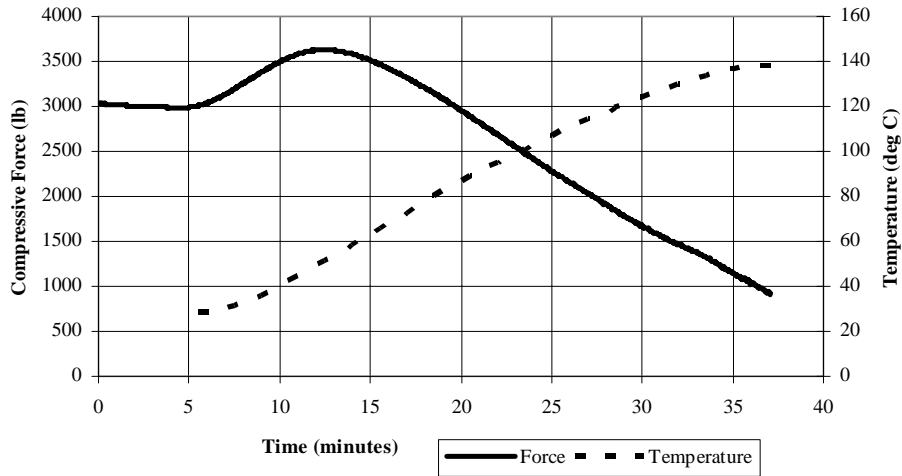


Figure 4.3: Displacement controlled hot pressing for initial 5" x 1" samples.

Table 4.3: Mechanical properties of initial bend samples.

Test #	ρ g/cm ³	Brinell		
		Hardness kg/mm ²	E psi	σ psi
6	0.334	<0.24	5.49E+04	151
7	0.382	<0.24	6.98E+04	190
8	0.406	<0.24	1.22E+05	292
9	0.579	0.95	3.61E+05	539
5	0.806	3.16	7.19E+05	1627
10	0.837	2.61	2.54E+06	2217
3	0.989	6.5	3.10E+06	4902
4	1.034		2.49E+06	4455

decrease in viscosity, shrinkage, mechanical hardening before cooling and irreversible particle bonding.

After the displacement control tests were analyzed, we suspected that the initial thermoplastic assumption was wrong. This greatly reduced the usefulness of the final temperature-pressure combinations, but the correlations between density and mechanical properties were still deemed valuable. So, an alternate experiment to confirm the thermosetting properties was performed.

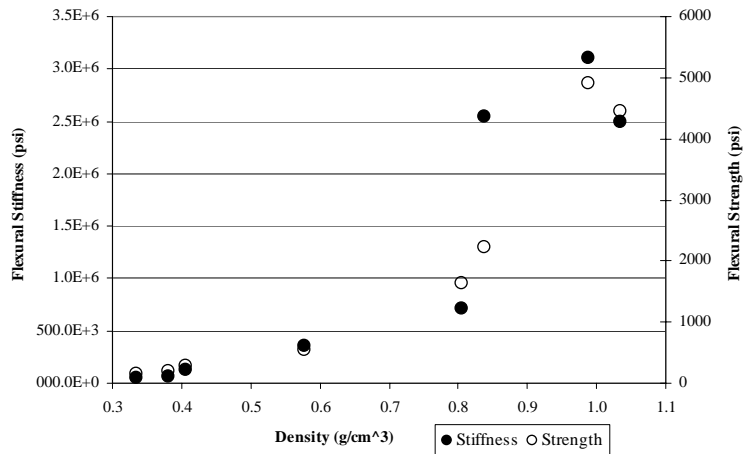


Figure 4.4: Flexural stiffness and strength vs. density for initial board samples.

4.2 Confirmation of Thermosetting Properties

For confirmation of the polymeric structure, a two part experiment was performed. In test 1, pith hand packed into a die was heated to 160°C, cooled to 100°C and then compressed to 0.25". For test 2, an identical pith amount was heated to 100°C and then compressed to 0.25". If the pith behaves as a thermoplastic, the results would be identical. Heating to 160°C would have no effect on the final combination of 100°C and 0.25" thick. However, if the pith is a thermoset, the heating to 160°C would advance an irreversible reaction in the temperature range of 130°C - 160°C (see Figure 4.1). Cooling to 100°C and compressing to 0.25" thick would break any bonds formed due to the chemical reactions at the higher temperature, making the pith brittle.

Test 1 resulted in a compressed block that, after relaxing overnight, broke under its own weight. It was only particles compressed together without any inter-particle bonding. Test 2 resulted in a solid piece that, while relatively weak due to the low cure temperature, was well bonded together. This confirms that the pith

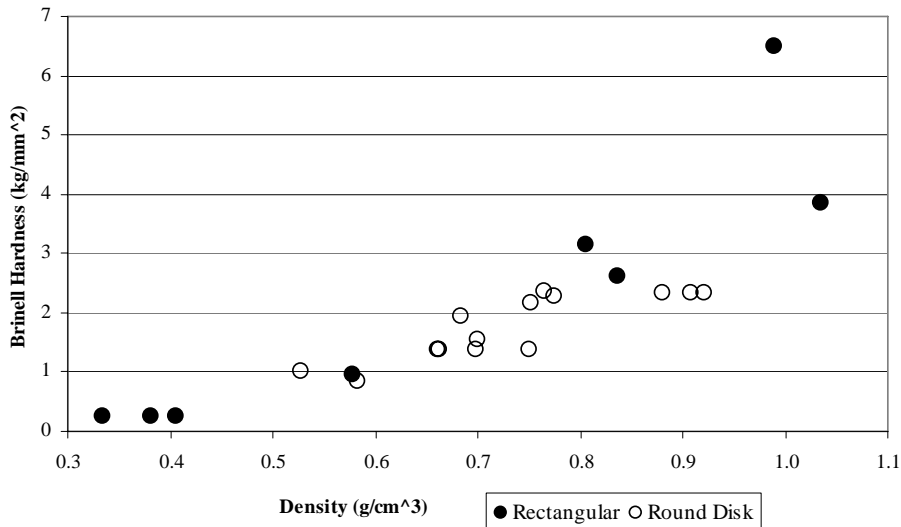


Figure 4.5: Brinell hardness vs. density for all initial samples.

does indeed behave as a thermosetting polymer as indicated by van Dam in the literature. If the pith was a thermoplastic, there would have been no difference in the results since the physical changes would have been totally reversible. However, because the pith is a thermoset, it chemically reacted under very low pressure during the initial heating, and any bonds formed were broken during compression.

Our alternate experiment confirms that the pith does indeed behave as a thermosetting polymer, but is also raised the question of why the DSC scans performed at Texas A & M did not show the exotherm. Later, at the meeting where we acquired their final report, van Dam explained that their successful scan had been on compacted pith and that they also saw no exotherm using only loose pith.

4.3 Displacement Control Experiments

Once we were sure that the pith did indeed behave as a thermoset, we decided to proceed with future experiments under displacement control. There were several reasons for this decision. Previous experiments indicated that properties were associated mostly with density which is most easily controlled by displacement control processing. Also, the available version of TestWorks software had no automatic load

control function. The later purchase of new software let us perform additional experiments with load control. The experiments run in displacement control and what was learned from them will be described next.

Displacement controlled hot pressing experiments were performed to answer critical questions about the effects of hot pressing temperature, pressure path and moisture changes on the mechanical properties of binderless particleboard. These various displacement hot pressing experiments were performed on pith acquired from compressed garden blocks.

4.3.1 Effect of Moisture Content

Mechanical testing results from samples produced with varying initial moisture contents are presented in Table 4.4. The results show that mechanical properties are only associated by density and not necessarily by moisture content. That is, the mechanical results increase monotonically with the densities. Although there is no direct relationship between and mechanical properties and initial moisture content, the moisture content can be a great asset in achieving the desired density.

Table 4.4: Results from experiments on the effects of moisture content [8].

Water %	ρ g/cm ³	E psi	σ psi
15.4	0.9	4.36E+05	5,923
15	0.77	1.64E+05	3,200
5.2	0.78	1.48E+05	2,327

The pith relaxes viscoelastically under the high load of initial displacement. As seen in Figure 4.7, when the heat is applied, the compressive load on the pith with the lower moisture content behaves similarly to experiment in Figure 4.3 where the load increased before decreasing. The compressive load on the pith with the higher moisture content decreased immediately when the heat was added. This behavior is typical of wood products. The added heat has a twofold effect. The thermal

expansion of compressed wood raises the load and the loss of internal water from the elevated temperature allows increased contraction in response to the displacement. Each of these competing factors overshadows the other at different points in the processing. The initial thermal expansion increases the load until the wood loses enough water for the contraction to overcome the expansion, at which time the compressive load drops to a new equilibrium. The pith with the higher moisture content loses moisture at a higher rate so its thermal expansion never has a chance to raise the compressive load [3].

This effect can be observed repeatedly when the temperature is raised incrementally below the cure temperature of the pith. An example of this is shown in Figure 4.6. With each temperature rise in Figure 4.6, the load increases slightly before dropping to a new equilibrium load.

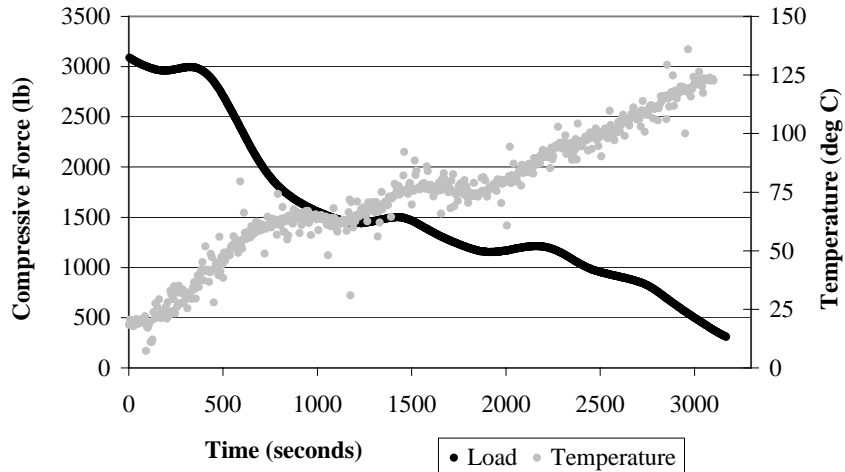


Figure 4.6: Compressive load response to pre-cure temperature changes.

The moisture content also affects the compressive load at equilibrium and the time required to for the relaxation or shrinkage to overcomes the thermal expansion. Results from two experiments with different initial moisture content have been graphed together in Figure 4.7. The heat was added once the ambient temperature relaxation and shrinkage had stopped as indicated on Figure 4.7

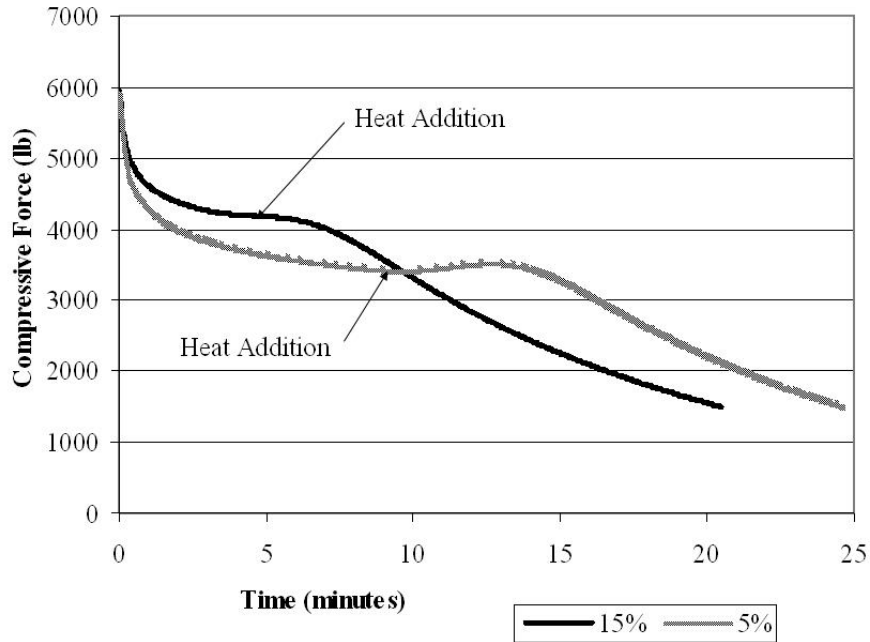


Figure 4.7: Load paths for compressed samples with different moisture contents.

Absorbed water can affect the pith in two ways. It acts as a plasticizer in helping the pith to flow, facilitating the packing and may also serve as a transport mechanism for the phenolic compounds inside the pith particles. Thus, a sufficient amount of water is necessary to help compact particles to increase density, but not so much water that blisters form from the rapid vaporization.

Our experimental findings on the effect of the initial moisture content in the pith were confirmed by subsequent results from van Dam's report. The effect of initial moisture content in the pith on the flexural stiffness and strength of the hot pressed pith can be found in Figure 4.8. The samples produced in our research had mechanical properties equal to those in Figure 4.8, but at a lower density. Van Dam also reported that high moisture contents resulted in blisters and warping and that low moisture content reduced the flow of the lignin which was noted above.

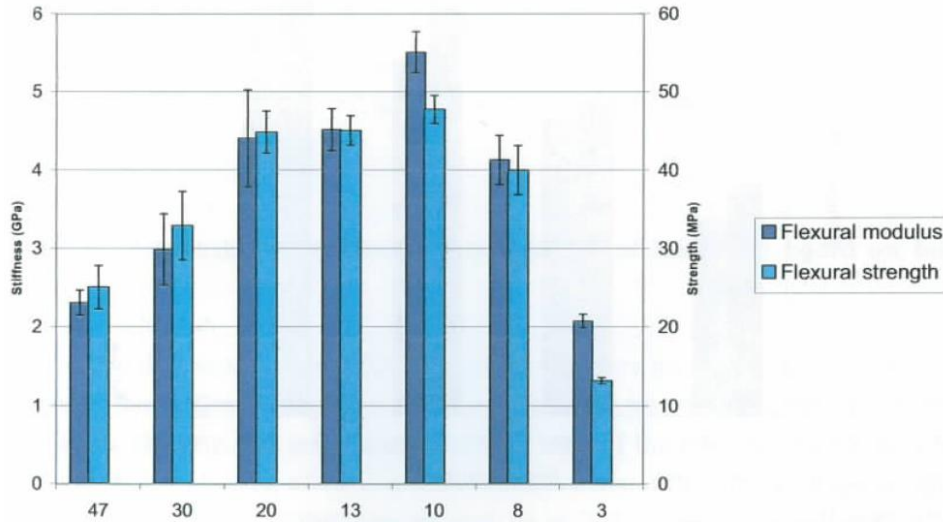


Figure 4.8: Effect of initial moisture content.

4.3.2 Property Dependence on Flow and Cure Temperatures

Time-Temperature-Transition TTT-diagrams present the curing behavior of a thermosetting resin. Figure 4.9 is a schematic TTT-diagram that illustrates the different curing stages for a typical thermosetting resin as a function of time and temperature. The pith does not exactly follow the behavior of other thermosetting resins since it is not a liquid. Most thermosetting resins can be poured into molds and cured with a curing agent at relatively low temperatures and almost no pressure. The coconut pith has its thermosetting properties bookended by its behavior as solid woody materials. This means that the only lines on the TTT-diagram worth considering are the full cure and char lines. Temperature and time combinations must be identified that give a full cure without starting thermal degradation. Since the time axis is logarithmic, the useful ranges are usually easier to find by varying time.

Samples were compressed at different temperatures to determine the temperature dependence of the pressure required to give a suitable density to facilitate curing. A lower pressures and therefore a lower total force will reduce the size and cost of the press required for hot pressing. The different curing temperatures were

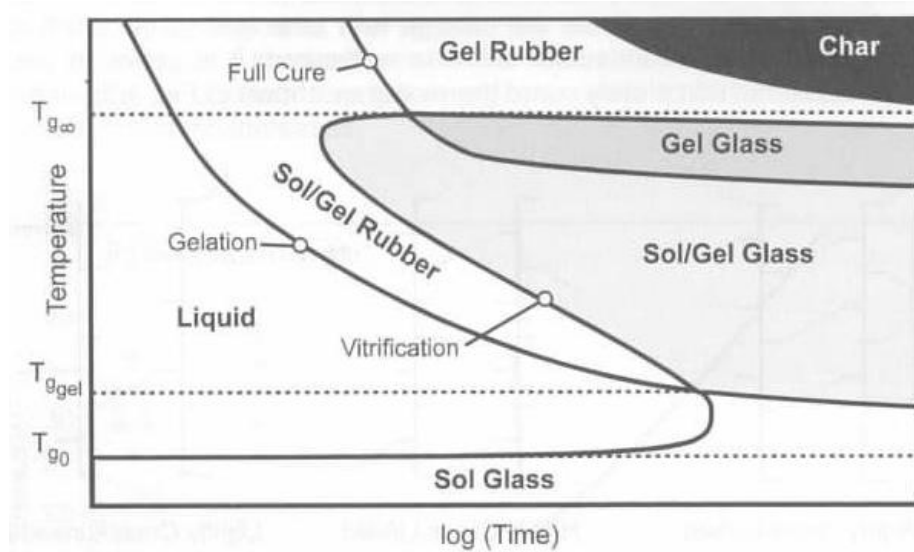


Figure 4.9: General time-temperature-transition diagram for general thermosetting polymer [9].

explored to determine the temperature range to give a full cure and the temperature above which the pith chars or degrades. The results are presented in Table 4.5.

Table 4.5: Effects of Flow and Cure Temperatures.

Compression Temperature °C	Compression Load lbf	Intended Cure Temperature °C	Density g/cm ³	E psi	σ psi
40	6600	140	0.66	NA	NA
105	2700	140	0.68	1.19E+05	2614
110	2300	160	0.66	1.19E+05	2224
120	2000	180	0.65	1.10E+05	2198

The compression load to achieve a density of 0.66 g/cm³ dropped from 6,600 lb to 2,500 lb when the press temperature was raised from 40°C to 100°C, which was expected from the relaxation temperatures previously observed. Smaller presses can be used if pith is compressed after it is heated to a temperature above the minimum flow but below the cure temperature in our laboratory. Because only cold mold processing with a slow temperature increases was available, all samples cured before reaching temperatures as high as those used by van Dam could be reached.

However, samples with better flexural strength for a given density were obtained when the maximum temperature to which the die was heated for curing was lower. The moduli data and densities in Table 4.5 wer essentially the same for maximum curing temperatures of 140°C , 160°C and 180°C, indicating that all samples werer equally cured to the same degree.

The load and temperature paths from one of these experiments are presented in 4.10. Much insights can be gained from an analysis of the results. In this test, the pith was compressed at 125°C so all of the moisture had been driven away before the load was applied. This means that the load drop seen from the time the displacement was applied at 15 minutes until 22 minutes was due only to pith viscoelastic relaxation and shrinkage. Until the minute 22, the processing behavior was dominated by the piths properties as a compacted but not yet cured biomass. At minute 22, the slope of the unloading path becomes steeper and begins an entirely new decay function. This change was from the shrinkage due to curing supplementing the relaxation of the pith. This showed the point at which the thermosetting properties begin to dominate the pith’s properties.

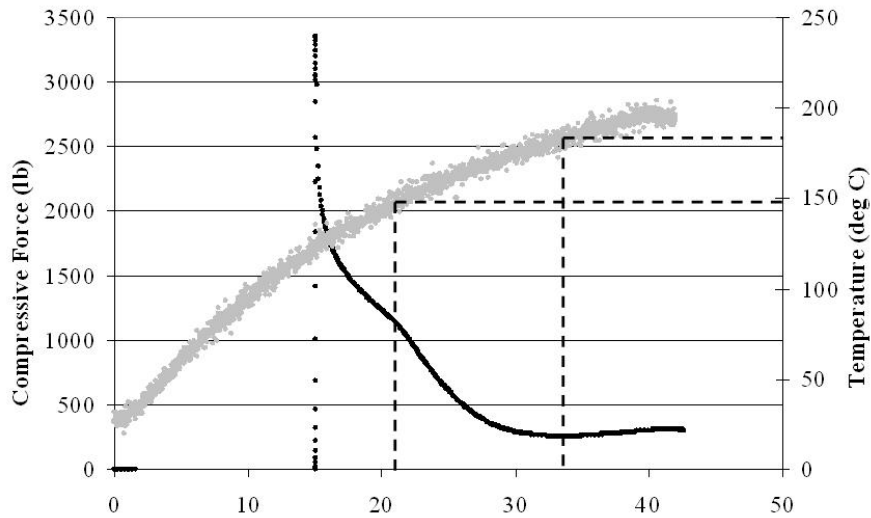


Figure 4.10: Load and temperature paths for compressed pith during processing.

This transition occurs near 140°C in Figure 4.10 and as soon as 130°C in similar experiments. The reaction is shown to be complete around 160°C-170°C, indicated by the line added at minute 35. This was where consolidation ended and the pith's properties as a solid wood piece began to dominate. After approximately 15 minutes, the same time stated in the literature, there was a rise in pressure from the thermal expansion of the newly formed solid wood piece.

This one test allowed us to identify the general locations of the full cure and char lines on a TTT-diagram for future processing. The discovered maximum temperature range of 160°C-170°C was in conflict with the results van Dam reported of 180°C presented in the literature, but this was later reconciled with a correction in the final report. The initial experimental conditions from the literature were determined with the temperature setting on the hot press. When van Dam took the work to industrial scale equipment, thermocouples were introduced and the temperature range was corrected to 160°C to 170°C [8].

This means that the pith fully cures at temperatures as low as 130°C, but may take a little longer to cure than it would at a higher temperature. The char region was reached quickly when the pith temperature was 160°C, which agrees with van Dam's later results.

4.4 Importance of Cross Link Density

Particle boards made 6" x 9" x 0.5" by using pith from compressed blocks with displacement controlled hot pressing on the Dake lab press. The mechanical properties of all of these boards were one the order of 10% percent of results obtained from previous samples similar densities. We hypothesized that the cross link density is an important factor in determining the mechanical properties and that the compression of the pith for shipping, the particle size distribution and the difference in load and displacement control may be important factors for cross linking density.

4.4.1 Cross linking

If all samples were fully cured and had the same density, the cross linking densities could still be drastically different, and different cross linking densities would be evident from major differences in different mechanical properties like flexural modulus and strength. A graphical explanation of the importance of cross linking density is presented in Figure 4.11. Without chemical cross-linking, only mechanical interlocking holds adjacent pith particles together and using pre-pressed pith probably reduces the degree of mechanical interlocking that can be achieved.

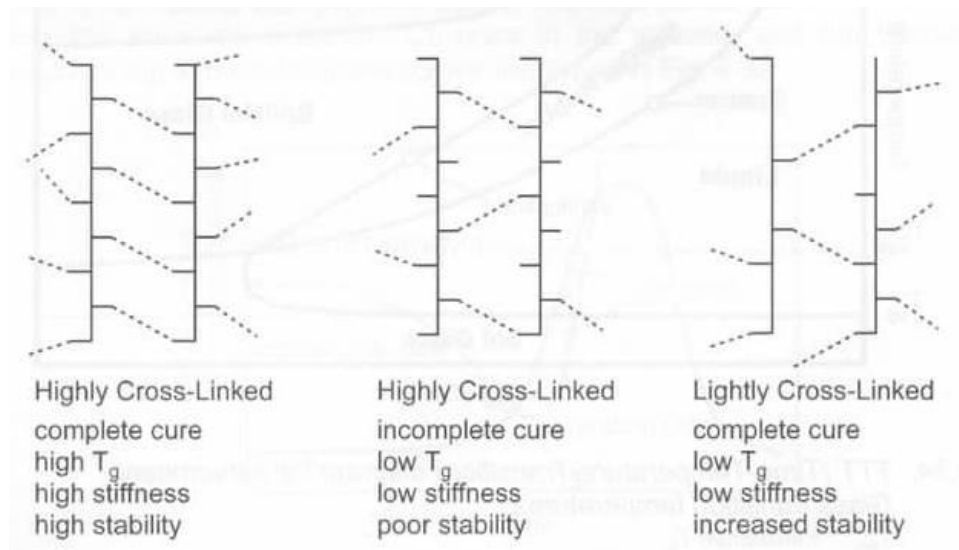


Figure 4.11: Diagram illustrating degree of cure in thermosets [9].

4.4.2 Effect of Particle Size

Coconut pith separated from the fiber has a known distribution of particle sizes. If differences in particle size affect packing efficiency, they could also affect cross-link density and therefore the mechanical properties. A distribution of particle sizes from chopped husks was presented in by van Dam, [8] but the effect of varying particle sizes on cocoboard properties was not measured.

Compressed coconut pith was grated and separated with three sieves with holes of sizes $250\mu\text{m}$, $600\mu\text{m}$, and 3.5mm . The particles that were too large for the

3.5mm sieve were discarded. The mechanical properties of samples of two different densities produced by displacement controlled hot pressing on the MTS fame are presented in Table 4.6.

Table 4.6: Effects of particle size.

sample size	ρ g/cm ³	E psi	σ psi	1000 σ /E
3.5 mm	0.46	1.07E+04	44	4.1
600 μ m	0.5	1.06E+04	40	3.8
250 μ m	0.52	1.01E+04	31	3.1
3.5 mm	0.81	4.52E+04	156	3.5
250 μ m	0.85	5.96E+04	154	2.6
600 μ m	0.87	6.82E+04	175	2.6

The particle sizes did not show any discernable relationships with the mechanical properties. As in previous experiments, the mechanical properties were positively correlated with density. The fiber volume fraction has a larger impact on the strength while stiffness is less effected by the pressure of fiber in the pith, which is consistent with common fiber reinforced composite theory.

4.4.3 Effects of Pith Compression

When coconut pith is sold for gardening applications, it is compressed into blocks for efficient shipping. Many of our experiments at Baylor were performed with uncompressed pith from these blocks due to the lack of available husks and limited milling capacity (because it was done by hand). The degree to which compression of the pith for shipping overseas from the Philippines to the U.S. affects the quality of particle board that can be produced was investigated.

A scanning electron microscope (SEM) was used to view the microstructures of coconut pith from raw husks from Mexico and from compressed blocks from the Philippines. It should be noted that the pith is compressed to 20% of its original volume to reduce shipping cost. The concern is that this severe compression might permanently deform the thin walled shell structure, affecting its behavior in hot

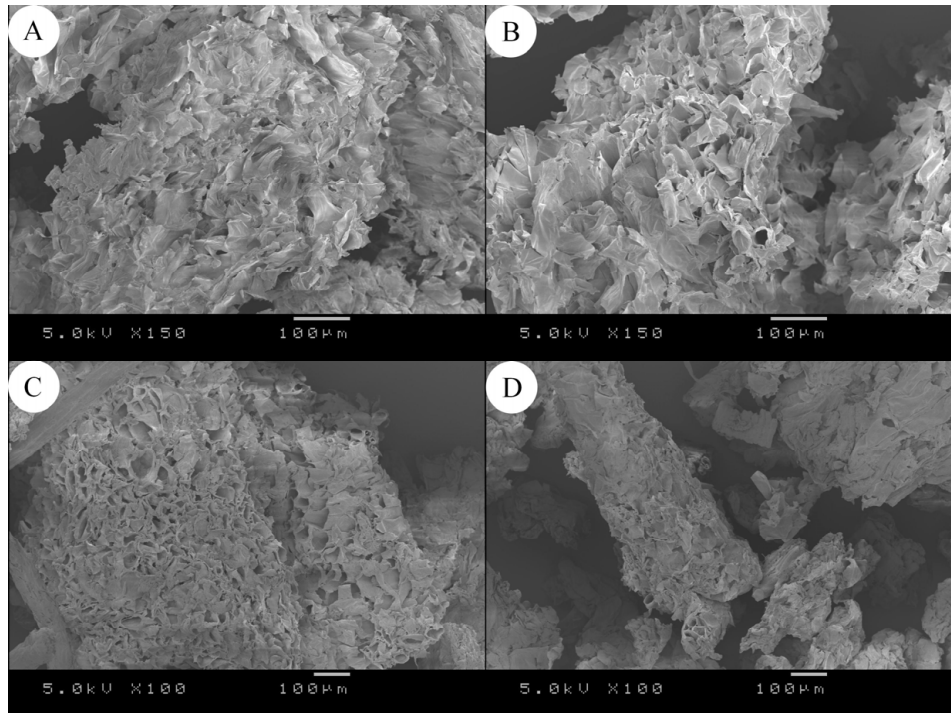


Figure 4.12: Micrographs of coconut pith from (A, B) raw husks and (C, D) compressed blocks.

pressing of its water retention in gardening. A sample of the many micrographs that were taken is presented in Figure 4.12.

The general sizes of pith particles were the same for both samples, but the surface textures were notably different. The raw pith had highly textured surfaces with many protruding edges. The compressed pith had most of its individual edges folded inward that created a smoother texture. Because pith particles can only bond where they touch, we thought that the less textured pith could have a decreased cross linking density and therefore lower properties.

4.4.4 *Effects of Load Control*

After reviewing the effect different processing variables on the resultant mechanical properties of hot pressed particle board, it appears that the experiments performed under displacement control give inferior stiffness and strength. The large

drops observed in mechanical properties across all densities for particle board made under displacement control can only be the result of insufficient chemical cross linking. The loss of cross-link density results from too little pressure to put molecules in sufficiently close proximity to chemically react.

4.4.5 Combined Experiment for Compression and Load Control

Samples were produced from bulk raw pith and compressed blocks under identical load and displacement control conditions. The results are presented in Table 4.7

Table 4.7: Effects of compression and load control.

Pith Source	Method	ρ g/cm ³	E psi	σ psi
raw	load control	1.21	2.32.E+05	466
compressed	load control	1.32	3.54.E+05	506
raw	displacement control	1.18	2.05.E+05	333

All three samples were in the same density and property ranges. This showed that neither the different sources of pith nor the choice between load and displacement control are important as long as a enough pressure is applied to get and keep the particles in close proximity before and during curing. This is most easily accomplished through load control, but displacement control can be used. If an initial overload is place on the pith so that the drop in load from relaxation stays above the required minimum pressure, then the results will be similar to those of a constant pressure above the required minimum pressure. We have observed the minimum pressure after over load to be as low as 200 psi, but the minimum value for load control has not yet been clearly established.

The strength values in Table 4.7 are very low compared to results of similar samples. The short fibers that were present in most samples that, as explained earlier provide most of the strength, were filtered out of the pith before the experiments

were performed. The results only confirmed that the mechanically separated pith provides essentially the same feed stock as the compressed blocks. There are still unanswered questions about the differences between hand and mechanical separation of the pith.

CHAPTER FIVE

Summary and Recommendations for Future Work

5.1 Summary of Results

- (1) The phenol molecules in the lignin portion of coconut pith have sufficient double covalent bonds and associated chemical reactivity to behave as a thermosetting resin, allowing the pith to be hot pressed into high quality particle board.
- (2) The pith's thermosetting behavior during processing is preceded by behavior as loosed woody material and followed by behavior as solid wood. Because of this unique combination of behaviors, the mechanical properties are controlled by the degree of cross-linking and density.
- (3) Degree of cross-linking density is a function of temperature path, pressure path and moisture content path. The cross-linking density is affected by the proximity of the pith particles, the availability of steam to distribute the increased temperature, and the combination of temperature and time to transition to a fully cured resin. Once the curing reaction is complete, the cured pith behaves like a solid piece of wood where, for a given cross-linking density, the Brinell hardness, flexural modulus, and flexural strength of the hot-pressed particle board increase with increasing density. An increase in the degree of cross-linking will give increased mechanical properties at lower densities.
- (4) Initial moisture content has several effects on the pith processing. Too much moisture causes blistering from rapid vaporization and limits the density that can be achieved during pressing due too excessive physical displacement of

the pith by the excess water present. Too little water inhibits viscoplastic flow of the pith during pressing, which limits the density that can be achieved and the degree of cross linking that results, due to lack of proximity between the reacting molecules. A moisture content of 15% in the pith was found to give the highest density, and therefore, the best flexural modulus and flexural strength.

- (5) Temperature is a critical variable in hot pressing to achieve both the necessary flow and chemical cross linking to produce particle board with excellent mechanical properties. The temperature of the pith must exceed 130°C for the chemical cross linking reactions to occur, but to minimize time in the press, temperatures of 140°C to 160°C are recommended. Using a temperature higher than 160°C will risk damaging the finish of the particle board due to blistering or charring. The temperatures mentioned above are the temperatures at the center of the pith, not the temperatures of heating elements, which will be somewhat higher.
- (6) Pressure is required to keep pith particle in close enough proximity that the phenol molecules can bond together. The proximity can be from either high initial displacement loads that remain high enough to keep a minimum pressure or from moderate constant loads. Pith sources may have short fibers incorporated in their mixture. These fibers affect the fiber volume fraction and the flexural strength of the particleboard. The flexural stiffness is the best indicator of the quality of processing of the pith since it behaves as the base matrix of the pith/fiber composite.
- (7) Figure 5.1 shows the relationships between flexural stiffness and density for the produced Cocoboard and other wood product options.

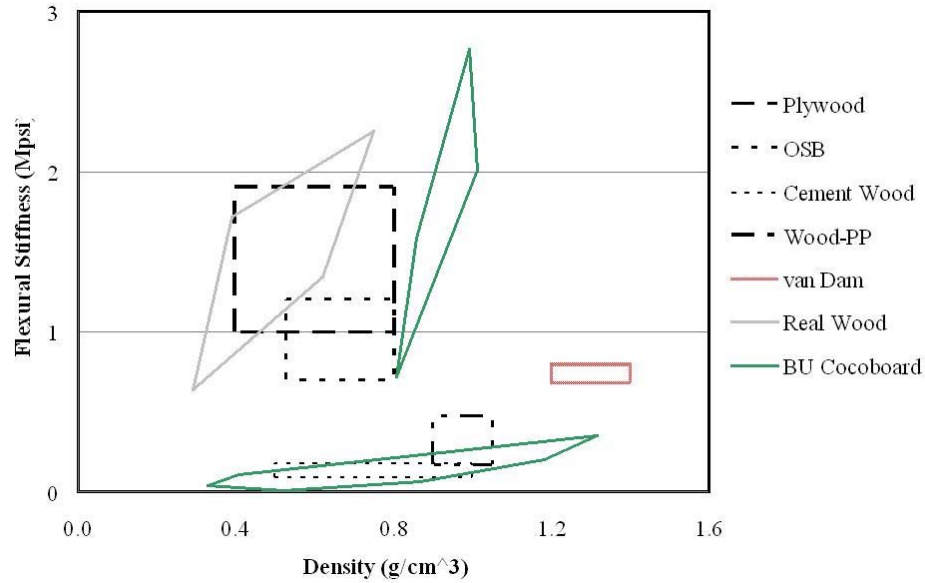


Figure 5.1: Stiffness vs. density comparison for wood composites [3].

5.2 Recommendations for Future Work

- (1) The relationships and interactions between temperature path, pressure path and moisture content path are complicated and little understood yet. Future research should be focused on maximizing cross-link density so that superior quality, lightweight boards may be produced like the high stiffness grouping of BU cocoboard in Figure 5.1.
- (2) Those excellent results, obtained with a unique combination of material and processing conditions, were very difficult to repeat with the available equipment. Any additional work should start with an attempt to repeat the best results from this research with the newly acquired equipment and include a study on the morphological differences between the hand separated raw pith and the machine separated raw pith.

APPENDIX

APPENDIX A

Results from CFC/FIGHF/11 and Preceding Publications

The behavior of coconut pith during hot pressing is complicated. Each variable can affect the final product, but effects are not necessarily independent. The first step to having a complete understanding of processing behavior is to examine each variable separately. This requires a standard production process, often chosen through initial ad hoc experimentation, from which to deviate to determine the effects of each variable. The first level of experimentation allows irrelevant variables to be eliminated from consideration, categorical variables to be optimized with a fixed choice, and probable dependant variables to be identified for further optimization in future investigations.

Not every possible variable was considered and tested by CFC/FIGHF/11 and its prior published work. This was due to certain assumptions made about processing conditions and a lack of material processing experience. This Appendix sorts important results from CFC/FIGHF/11 by processing variable and discusses the conclusions about each one by identifying unimportant variables, selecting preferred categorical choices, and choosing interrelated variables for further research.

A.1 Standard Production Process

In order to accurately determine the effect of each variable, a standard processing method must be established as a control. CFC/FIGHF/11 uses processing conditions decided on during the development of its preliminary papers. In this initial research, hot pressing was performed in the temperature range 100-250°C and the pressure range 80-200 bar during 10-60 minutes. The best mechanical properties were obtained for dry-milled husk applying 15 minutes 150 bar pressure at 180°C.

All subsequent lab scale experiments were performed with this processing procedure. The board samples were cut and subjected to mechanical testing. Any additional testing is noted in the following section [8].

Dry-milled husk material was hot pressed with different combinations of temperature (100-250°C), pressure (80-200 bar), and time (10-60 minutes). The moisture content was known to have a great influence on the hot pressing technique, but was considered separately as an independent variable. At least for higher scaled production, the pressing load remained until the cocoboard cooled to 30°C.

A.2 Raw Husk Type

Coconuts of different ages and varieties may affect cocoboard properties, so coconut raw material properties were analyzed in terms of their morphological and chemical features. es may have different internal structures and chemical compositions that could affect cocoboard properties, so six species from The Philippines region were compared for the CFC/FIGHF/11 project.

No significant differences between husks from different coconut varieties were observed. Also, scanning electron micrographs, which show detailed pictures of fiber and pith morphology, did not show significant differences in husk appearances between the coconut varieties investigated [8]. The chemical compositions of the investigated varieties were the same, and the total lignin contents were all within a 4% range. Since only slight differences were observed between mature coconut husks of different origins, all are suitable as input feedstock for cocoboard production [7].

Boards were produced from all six varieties, and the stiffness values ranged from 4.8 to 5.3 GPA (0.70-0.77 Mpsi), and strength from 43 to 53 MPa (6.2-7.7 ksi). All of the cocoboards were fully suitable for common woodworking procedures, and in terms of performance, at least equaled MDF and outperformed particle board, but the densities were nearly twice as much at 1.4g/cm³ [8].

While the chemical composition and morphology coconut fiber was investigated and included in the cocoboard feedstock along with the pith, only the pith results were considered since the pith behavior controls the base properties as the composite matrix. A consideration of the fiber volume fraction should be performed independently, similar to the production of other fiber reinforced composites.

Even though all but one of the investigated varieties was from The Philippines, it is reasonable to assume that all other varieties will have the same results. So, husk variety were not be considered an important processing variable. If, in the future, it is discovered that some varieties produce a deviation in properties or are not suitable raw material at all, this should be considered a site selection factor rather than a production factor.

A.3 Raw Husk Age

As young green coconuts mature to brown coconuts, their internal structure changes greatly. Initially, the shell and husk are a continuous mass of soft pulpy wood. As the coconut matures, the wood separates into the hard shell and the thick fibrous husk. Five maturity levels (6, 7, 9, 11, 13 months) were investigated to find the effect of age on the cocoboard properties.

The amount of husk and the dry husk fraction of the nut increased with age [8]. No comparisons of the macro or micro structure were made based on age, but the metamorphosis of the husk content is commonly understood.

A change in chemical composition of husks was observed when comparing nuts of different maturity. The amount of extractives decreased strongly during maturing of the nut, while the amount of lignin and carbohydrates increased. A gradual increase of glucose was observed, associated with formation of cellulose, while for other sugars no dramatic change in the relative amounts was observed as function of maturity [7].

As part of the chemical analysis, the weight loss as a function of temperature was measured. Representations of the results can be seen in Figures A.1 and A.2.

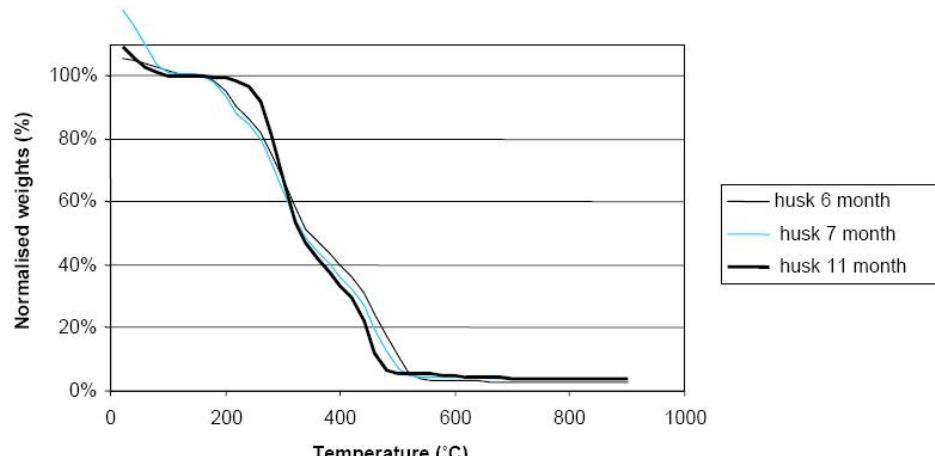


Figure A.1: Thermogravimetric analysis (TGA) of LAGT coconut husks of different maturity (6, 7 and 11 months) [8].

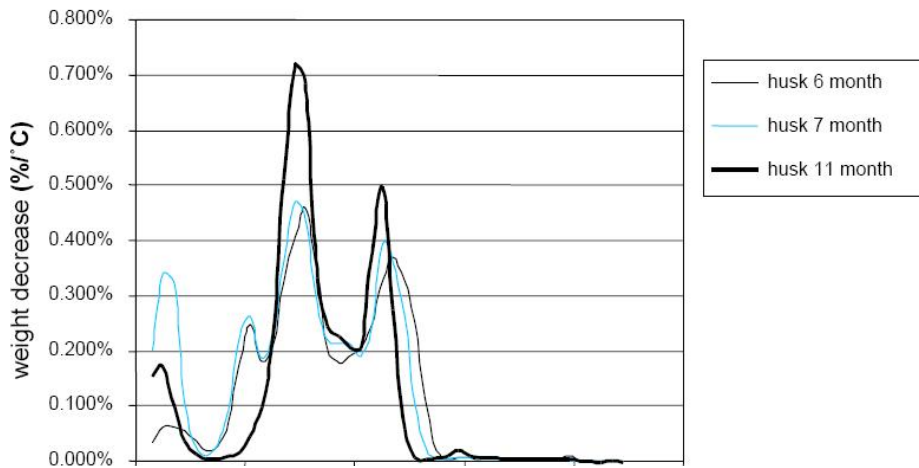


Figure A.2: Weight decrease (%/C) as a function of temperature [8].

Boards were produced with the standard processing method. Samples were made from 6, 7, and 11 month husks and from 6 and 7 month husk/shell mixtures. The results were graphed and tabulated in Figure A.3 and Table A.1 respectively.

Eleven month cocoboard outperformed MDF, but at nearly twice the density. It also outperformed all younger cocoboard feedstocks. The presence of shell in

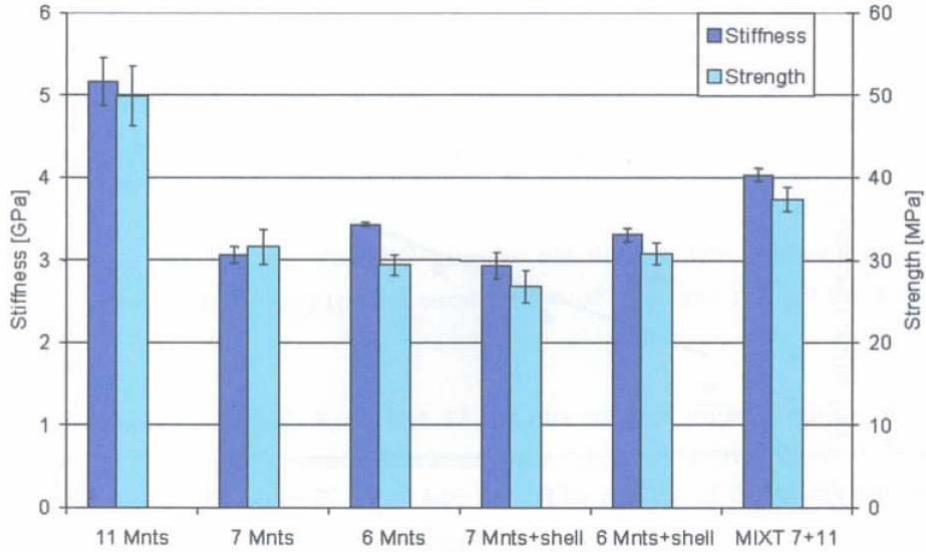


Figure A.3: Stiffness and strength of cocoboards from different aged husks [8].

Table A.1: Mechanical properties of cocoboards made from different aged husks [8].

Age months	E psi	σ psi	ρ g/cm ³
11	725E+3	7.3E+3	1.4
6 and 7	450E+3	4.5E+3	1.4
MDF	450E+3	1.9E+3	0.8

young samples had no effect on mechanical properties [8]. The loss of extractives in the maturity of the 11 month husks can be seen in Figures A.2 and A.3 at 200°C by the sudden drop in weight in only the younger husks.

In Figure A.1, the separation between the 11 month husk and the 6 and 7 month husk at 200°C indicates that the extractives are expelled starting at 150°C. The improvement in properties would be expected with more advanced age since it is the lignin, found more in the mature husks, that gives the strengthening thermosetting behavior. The immature husks would be a possible feed stock, but will not be considered since the mature nuts have more available material with higher lignin content. This removes the age of the coconut as a variable in the production model.

A.4 Husk Milling Method

Three methods for the separation of pith and fiber were evaluated: steam explosion, extrusion and milling. Boards were made from each sample using the standard procedure and the mechanical and physical properties were compared. All three methods separated the husk into small particles and short fibers that were appropriate feedstock.

The three methods were compared based on energy requirement, equipment investment and quality of product. Milling bested steam explosion and extrusion in all three categories [8]. Since the best results were obtained with milling, it was selected for all further studies [6].

Since milling is clearly the preferred method of separation, the method used will not be a variable included in future processing models. Before results from the CFC report were received, it had already decided milling is the preferred method based on equipment costs and interviews with Pilipino coconut co-op managers.

A.5 Processing Conditions

The moisture content was found to have a great influence on the hot pressing technique, but was considered as an independent variable. Boards were made with various water contents with the standard production procedure.

The reproducible conditions with the best mechanical properties were obtained with 150 bar pressure at 180°C for 15 minutes [8]. These conditions were used for all experiments. Cold molds, if heated longer, were shown to be as effective as hot molds, but no details such as temperature and load path were given on the T-P-t experiment results that would give insight into the processing behavior. It was stated that load control was necessary over displacement control, but no experimental results or reasoning was presented. No experimental comparisons or reasoning was given for the continued load while cooling.

High moisture content resulted in blisters and warping while very low moisture content reduced mechanical properties. Water is pressed out of the husk when the moisture content is above 35%, and the board properties were superior when the moisture content was at 10%. Mechanical test results were graphed and can be found in Figure A.4

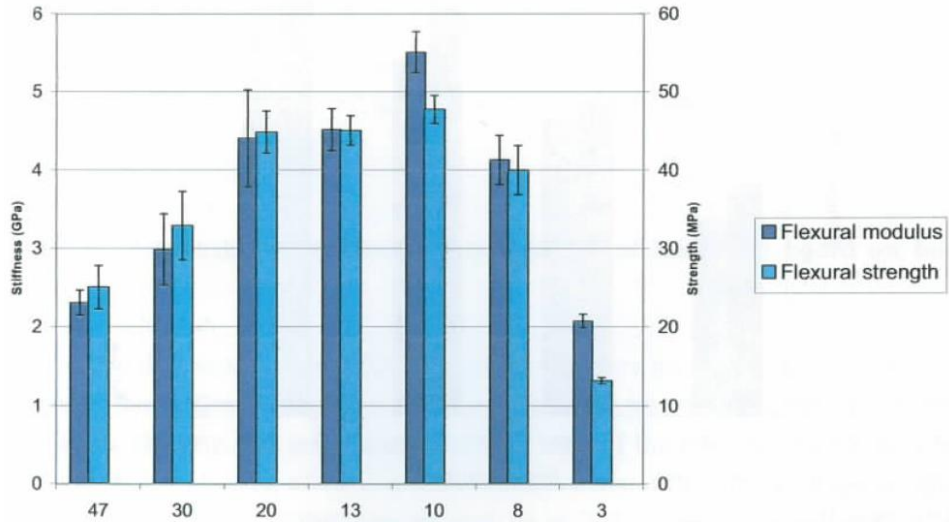


Figure A.4: Influence of initial moisture on the boards' mechanical properties [8].

It was recommended that moisture content be kept below 12% to prevent blistering. Although, moisture content may be eliminated as a future variable, assuming blister prevention is a priority, there is no mention to the phenomenological process driving the effect of moisture. A good understanding of what is happening chemically is important to gaining a fuller understanding.

A.6 Post Processing

Because the pith moisture content before processing influenced the board properties, it was believed that the board moisture content would also affect mechanical properties. Boards made from husks conditioned at 50% and 95% relative humidity were each conditioned at 50% and 95% relative humidity. The results were tabulated and can be found in Table A.2.

When the boards made from 50% RH husk were conditioned at 95% there was considerable fungal growth. When the boards made from 95% husk were conditioned at 50% considerable warping occurred. The mechanical properties were considerably less for the board made from the husk at 95% RH. It was suggested that than excess of water in the husk hinders compression and good interaction of the fiber and pith material [8].

Table A.2: Effects of post processing conditioning [8].

RH %	Strength Mpa	Modulus Gpa	Density g/cm ³
50	46 (2)	4.2 (0.1)	1.38 (0.01)
95	13 (1)	0.85 (0.02)	1.21 (0.01)

The husk conditioned at 95% humidity had a moisture content of 40%, so the results are consistent with previous moisture content tests. While it may be the case that initial moisture caused the drop in properties at the higher RH, it seems more likely that the additional moisture present in the finished board acted as a plasticizer in the 95% RH board. This is typical for polymers, which the lignin has shown to be, so the 95% RH boards could be dried for much improved performance.

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