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FACTORS AFFECTING WOOD PELLET DURABILITY

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by
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Abstract

Densification increases the bulk density of biomass, thereby increasing the efficiency of its transport and improving its competitiveness with low-cost fossil energy. The method of densification investigated herein is pelleting. The research examines factors affecting the durability of fuel pellets produced from various tree species. Pellet producers often observe differences in pelleting behavior depending on species mix. By isolating tree species, differences within wood fibers can be observed.

The Pellet Fuels Institute recently revised standards for pellet quality. One of these quality metrics is pellet durability. According to the literature, the melting of lignin is responsible for the lignin solid bridges that largely contribute to pellet durability. By producing pellets from tree species displaying a range of lignin contents and reproducing these lignin contents by mixing two feedstocks, the effect of lignin on pellet durability is isolated.

No impact on pellet durability due to lignin content is observed. In the absence of a clear lignin effect, bulk material properties of individual species are investigated. While lignin content varies within tree species, it was not observed to have an impact on pellet durability; however, significant differences in bulk material properties between pellet feedstocks that produced the highest and lowest durability values are observed. By looking at the bulk modulus and elastic response of densification feedstocks, among other bulk material properties, the biomass densification industry may gain insight into densification behavior. This is of particular importance as the industry looks to expand to feedstocks beyond woody biomass.

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Chapter 1

Introduction

With the recent spike in global energy prices, growing concern over climate change, and the push for energy independence, alternative means of energy production become increasingly viable. These alternatives include nuclear, aeolic, hydroelectric, photovoltaic, solar thermal, and biorenewable energy technologies. Biorenewable technologies are of particular interest to the agricultural sector as they will have a significant impact on the utilization of the nation's cropland. Here, the term biorenewable technologies refers to the direct combustion of biomass in addition to biofuel production from biomass.

Biomass has not been widely utilized due to its relatively low energy density when compared with fossil fuels. This low energy density results in inhibitive transportation costs and inconvenient storage and handling. To overcome these obstacles, biomass can be densified into a form that can be stored and handled in a manner consistent with that already developed for grains (Fasina and Sokhansanj, 1996). The most common forms of densified biomass are pellets, briquettes, and cubes.

Currently, wood pellets are produced for utilization in residential pellet stoves. Substantial opportunity exists for the utilization of other forms of biomass as a feedstock for pellet production, but the current market consists almost solely of wood pellets. Wood pellets are currently graded by the United States Pellet Fuel Institute (PFI). Recent adoption of new standards calls for producers to meet a minimum durability requirement. While some producers routinely meet this requirement, still others often fall short (C. Wiberg, personal communication, Sept. 2008).

Kaliyan and Morey (2006) define pellet quality in terms of the strength and durability of the pellets. Strength refers to both the impact and compressive resistance. Durability is a measure of the friability of the pellets. There are several established methods originally developed by the feed industry for the evaluation of these qualities that can be applied to the use of pellets in energy systems. These methods were designed to simulate the forces induced on the pellets in the storage and handling process. Test results are used as indicators of the level of breakage in storage and handling as pellets are transported from production to utilization. Of the quality metrics available for pellet testing, durability is most representative of the forces in transportation, storage, and handling, and is the only physical quality metric considered by industry.

Pellet quality is largely a function of the type of feedstock and feedstock and process parameters. The focus of this work is the effect of the variation of feedstock parameters on wood pellet durability. Through conversations with the PFI standards committee, tree species was identified as a critical variable in the wood pelleting industry. Due to high transportation costs associated with wood residues and roundwood¹, pellet producers are limited to a relatively small geographic radius from which to draw feedstock. Since tree species is largely a function of climatic and geologic conditions, producers are limited in their species selection. Along with tree species, moisture content was selected as an experimental variable. Moisture has long been identified as a key parameter in densification processes. By investigating the pelleting behavior of various species over a range of moisture contents, differences between tree species can be isolated, shedding additional light on the pelleting process.

¹ Whole logs

Chapter 2

Literature Review

The literature review addresses four major topics. First, it differentiates between various densification processes, identifying their strengths and drawbacks. Next, it discusses the mechanical interactions associated with densification processes with the goal of identifying those that will change with tree species. Following the mechanics discussion, the review addresses the parameters identified as affecting the durability and strength of densified products, including moisture content, particle size, chemical constituents of the feedstock, and densification process variables. Finally, the literature review concludes with a discussion on the quantification of strength and durability that provides definitions for both terms and presents methods for determining each.

2.1 Densification Processes

Densification processes can be broadly categorized by two methods: closed and open die compaction. Closed die compaction is commonly referred to as tableting or roll-press briquetting. The most common tableted products are pharmaceutical tablets and charcoal briquettes. By contrast, wood pellets are formed via extrusion through an open die. The extrusion process is a continuous process, constantly forcing feedstock through the die opening to form the densified product. The three most common forms of extruded biomass products are pellets, briquettes, and cubes.

2.1.1 Pellets

Pelleting has traditionally been employed in the processing of animal feeds to increase bulk density and promote their delivery in feed rations. Recently, high fossil fuel prices have led to

increased production of pellets for home heating. Currently, wood pellets dominate the marketplace, but alternative feedstocks are being investigated. Pellets are cylindrical in shape and typically have diameters ranging from 4.8-19 mm and lengths from 12.7 to 25.4 mm (Kaliyan and Morey, 2006). Pelleting is conducted using a pellet press, also called a pellet mill or pelletizer. Large-scale pellet mills consist of a die with an annular matrix of extrusion holes and a roller. Smaller, lab-scale units and a mobile unit recently developed by Buskirk Engineering (Ossian, IN) employ a horizontally configured die with vertical rollers; however, the principle is the same. The roller forces the feedstock through the die holes in the form of pellets where they are cut by a knife to the desired length and collected below the equipment. Typical production rates for industrial pellet mills are approximately 3.6 Mg/h.

2.1.2 Cubes

The cubing process was originally developed for the economical transport of forages, namely alfalfa, over long distances. Western Canada continues to enjoy a large cubing industry for the export of alfalfa. Cubes are rectangular boxes in shape and larger than pellets. Two types of cubes exist: micro-cubes and cubes. Micro-cube dimensions are approximately 16x16x32 mm and have densities of near 550 kg/m³. Cubes are larger, about 32x32x64 mm, and have lower densities (~450 kg/m³). A cuber consists of a single roller, auger, and die ring. Similar to the pellet mill, the roller forces the feedstock through square die openings in the ring. The cubes are then fed into a dryer/cooler and dried to a moisture content between 10-12% before moving on to storage. Sokhansanj and Turhollow (2004) report a cuber production rate of 4.7 Mg/h; however, it is noted that this is a reported production rate for hay, and does not necessarily extend to other forms of biomass.

2.1.3 Briquetting

Biomass briquettes are not to be confused with their charcoal relatives. Charcoal briquettes are the product of a tableting process using a roll press briquetter, whereas biomass briquettes are formed via an extrusion process. In biomass briquetting, the feedstock is forced through a heated die using a piston. Following compaction, briquettes are cooled in-line by moving via mass flow through a chute exposed to the ambient air. Briquetters may be mechanically or hydraulically driven, although the mechanical models consume less energy due to the substantial heat rejection associated with hydraulic power. Briquettes range from approximately 6.4-8.9 cm in diameter and can be cut to the desired length as they exit the cooling chute. Briquettes are also commonly referred to as “pucks” or “logs.” Pucks, as the name indicates, have the same general shape and dimensions as a hockey puck. Logs are of similar diameter, but greater length. Production rates for typical briquetters range from 0.9-1.8 Mg/h.

2.2 Densification Mechanics

An understanding of densification mechanics is necessary in the characterization of pellet quality to identify the mechanisms by which bonding occurs. The nature of particle interactions in densification processes was established by Rumpf (1962) and has been quoted by numerous researchers.

2.2.1 Solid Bridges

High temperatures experienced in densification processes cause the partial melting of feedstock constituents. The melting of these constituents facilitates molecular diffusion between particles at heightened temperatures. As the densified product cools, the melted constituents solidify, forming solid bridges. These solid bridges largely determine the strength of the final products (Manickam, 2003).

2.2.2 Attractive Forces Between Solid Particles

As the feedstock is compressed and inter-particle distance is reduced, intermolecular attractive forces play a role in the bonding of particles. These forces include valence attractions (electron sharing), hydrogen bonds, and van der Waals forces. Of these, van der Waals forces are thought to be the greatest contributor to attractive forces between particles. Attractive forces decrease significantly as particle size increases (Kaliyan and Morey, 2006).

2.2.3 Mechanical Interlocking

Mechanical interlocking may occur during the compression of fibrous, flat-shaped and bulky particles. It is thought to be a minor contributor to overall strength. Gray (1968) notes that mechanical interlocking may provide sufficient mechanical strength to resist disruptive forces caused by elastic recovery following compression.

2.2.4 Interfacial Forces and Capillary Pressure

Interfacial forces and capillary pressure are created by three distinct states that occur during the densification process. They are the pendular, funicular, and capillary states. The pendular state is characterized by an initial air gap between particles. As densification proceeds, these air gaps fill with liquid and the particles experience surface tension and capillary pressure (suction) due to the presence of the liquid. The funicular state is characterized by the filling of all voids with liquid. The funicular state is an intermediary between the pendular and capillary states. Optimal feedstock moisture content is essential to the development of interfacial forces and capillary pressure. Too little moisture and the funicular state will not fully develop, too much and the incompressibility of water will adversely affect the agglomeration of particles. The capillary state is characterized by attractive forces between the liquid gas interfaces within the agglomerate (interfacial forces). Capillary pressure and interfacial forces create temporary but

strong bonds that disappear once the liquid evaporates. Still, they are essential to the development of cohesive forces discussed in section 2.2.5 below. Rumpf (1962) asserts that the “cohesive strength of the agglomerate is attributed to the forces exerted by the pendular bridges and capillary suction pressure.”

2.2.5 Adhesive and Cohesive Forces

Immobile adsorption layers can form strong bonds between adjacent particles. These sorption layers are thought to facilitate densification either by smoothing out surface roughness and increasing the inter-particle contact area or by decreasing the inter-particle distance and allowing intermolecular attractive forces to participate in the bonding mechanism. Adsorption contact area increases significantly when solid particles are subjected to high pressures, resulting in high bonding forces (Rumpf, 1962).

2.3 Factors Affecting Pellet Strength and Durability

A number of factors significantly impacts the strength and durability of pellets. Strength refers to the compressive and impact resistance of the pellet, whereas durability pertains to the friability, or abrasive resistance of a pellet. The parameters considered are: moisture content, particle size, preheating/steam conditioning, chemical composition of the feedstock, the addition of binders and/or lubricants, and process variables. Process variables include die dimensions, die speed, and gap between the roller and die.

2.3.1 Moisture Content²

Some water is necessary in the pelleting process for the development of intermolecular forces. However, too great a moisture content adversely affects pellet quality. Optimum feedstock

² Unless otherwise stated, all moisture content values are presented on a wet basis

moisture content of several biomass feedstocks has been investigated by numerous researchers. In their work with corn stover, Kaliyan and Morey (2005) found that increasing the moisture content from 10 to 15% (w.b.) increased durability from 62 to 84%. Li and Liu (2000) and Ohmberger and Thek (2004) postulate that the optimal moisture content for the densification of woody biomass is between 6 and 12% and 8 and 12%, respectively. The effect of moisture content on the durability of wheat straw wafers was investigated by O'dogherty and Wheeler (1984); these authors identified the optimal range of moisture content to be between 10 and 20%. Grass biomass mixed with 20% alfalfa has an optimal moisture content of 11%. Deviations from this value were found to have a significant impact on pellet quality (Srivistava et al., 1981). Optimal moisture contents reported in the literature span a broad range, even for the same plant species. What is clear is that the effect of moisture on the densification process is not well understood. In fact, Kaliyan and Morey (2009) state that moisture acts both as a binder and a lubricant.

2.3.2 Particle Size

Particle size, along with moisture content, is one of the most significant factors affecting overall pellet quality. Finer particle sizes generally correspond with greater pellet strength and durability as larger particles serve as fissure points (MacBain 1966). Several researchers observed that optimal pellet quality is achieved with a mixture of particle sizes due to increased inter-particle bonding (mechanical interlocking) and the elimination of inter-particle spaces (attractive and adhesive and cohesive forces) (Payne, 1978; Kaliyan and Morey, 2006a; Shaw, 2008).

Hammer milling is the most common form of particle size reduction for biomass feedstocks entering the pelleting process. Vest (1993) surveyed several U.S. feed mills and concluded that

hammer mill screen sizes of either 3.2 mm or 3.2 to 4.0 mm (4.0 on top, 3.2 on bottom) produced the highest quality pellets.

2.3.3 Preheating and Steam Conditioning

Kaliyan and Morey (2006) note that it is essential to provide heat and moisture to activate inherent binders in the feedstock. The activation of these inherent binders (lignin, proteins, and starches) promotes the formation of solid bridges, a primary mechanism of particle agglomeration. Additionally, heat and moisture facilitate the plastic deformation of particles, thereby increasing the inter-particle contact area and decreasing the inter-particle distance. Plastic deformation contributes to pellet quality by bringing the particles in closer contact, simultaneously increasing intermolecular bonding and allowing miscible constituents to flow together.

2.3.4 Lubricants and Binders

Some producers may add lubricants or binders to the feedstock to assist in pellet formation. The most common lubricant is vegetable oil, and is added to reduce friction between the die and feedstock. Generally, lubricants are added by hardwood pellet producers, presumably due to the fibrous nature of their feedstock. As a rule, softwood pellet producers do not require die lubricant.

Binders are added to improve the durability of the feedstock. Common binders in the feed industry include calcium lignosulfonate, colloids, bentonite, starches, proteins and calcium hydroxide (Pfof, 1964; Tabil and Sokhansanj, 1996). Binders are rarely used in the pelleting of biomass due to the associated additional cost.

2.3.5 Moisture, compressibility, and glass transition

As discussed previously, lignin solid bridges play a significant role in biomass densification. Conventional wisdom is that higher levels of lignin lead to a more durable pellet, since lignin acts as the “glue” that binds the product together. Lehtikangas (2001) reports a loose correlation between lignin content and pellet durability. Generally, longer storage in bulk leads to microbial degradation of the hemicelluloses and cellulose fractions of the feedstock, so that those that were stored longer have a larger percentage of lignin content.

As noted, moisture content also plays a critical role in pellet formation. However, the reason for this effect is not well understood. Traditional thinking is that some moisture content is necessary to develop intermolecular (van der Waals forces and hydrogen bonds) and interfacial forces that serve to bring particles in closer contact with one another during the binding process. However, it seems unlikely that these relatively small forces play such a large role in the densification mechanism. Perhaps a more plausible explanation is touched on by Kaliyan and Morey (2009) when they note the effect of moisture content on the glass transition temperature of corn stover and switchgrass. The authors report that increasing the moisture content of corn stover from 10 to 20 percent resulted in a decrease in the glass transition temperature. Similarly, in their work investigating the effect of glass transition temperature on microbial activity in processed foods, Chirfe and Del Pilar Buera (1994) observe that the glass transition temperatures of gluten, starch, and lignin fall dramatically with increases in moisture content. Given the large role of the formation of solid bridges in the densification process, it seems likely that the primary impact of increased moisture content is to lower the glass transition temperature of the constituents. The effect of moisture on the glass transition temperature of lignin, starch, and gluten is presented in Figure 2-1.

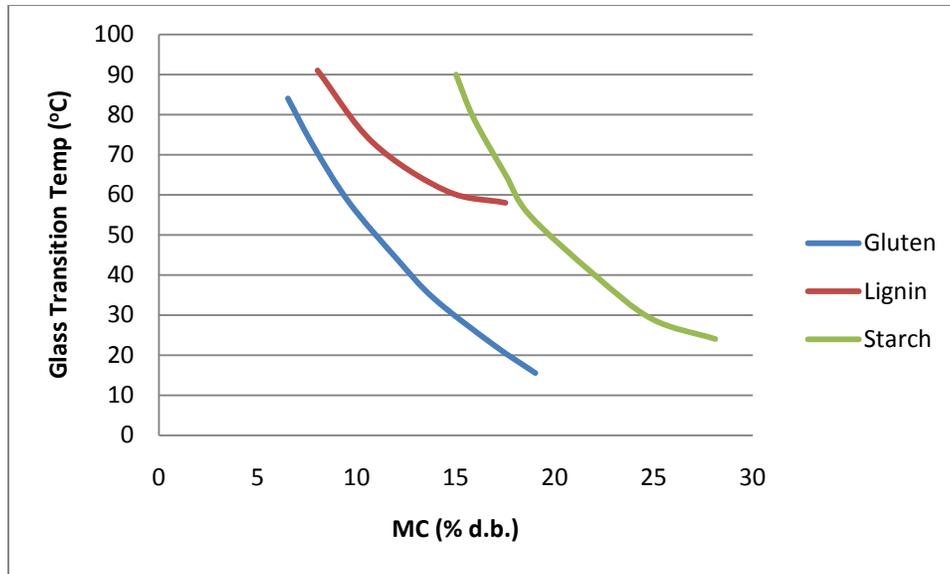


Figure 2-1 – Effect of moisture on glass transition temperature (recreated from Chirfe and Del Pilar Buera, 1994)

As can be seen from the figure above, the glass transition temperature, corresponding to the transition from the glassy to rubbery (hereafter referred to as elastic to plastic) regime of solids, drops rapidly as moisture content increases from 8 to 18 percent (d.b.). This coincides with the range of optimal moisture contents presented earlier in this section.

Glass transition temperature is measured via differential scanning calorimetry (DSC). DSC applies heat to two identical pans, one with material and one empty reference pan. The DSC adds heat to both pans to account for the heat capacity of the pan itself. Initially, the heat capacity of the sample will remain constant; however, as the material passes from the elastic into the plastic regime and the amorphous particles (lignin, in this case) begin to flow, the heat capacity of the sample will increase. This increase in heat capacity is also referred to as a second order transition. Eventually, the heat capacity of the sample stops increasing, corresponding to the end of the glass transition regime. The glass transition temperature is commonly taken to be

the temperature at the midpoint of the change in heat capacity of the sample (Kaliyan and Morey, 2006).

Further, as observed by Shaw (2009), feedstock compressibility has a large impact on compaction behavior. Some clarification of compressibility is necessary here. As referred to by Shaw, compressibility is the change in bulk density of a feedstock before and after loading. At the relatively high loading conditions investigated (~125 MPa), plastic deformation of particles is expected, as noted by Shaw. This is similar to the loading conditions experienced in commercial pellet mills, where plastic deformation plays a large role in pellet formation. Indeed, the plastic regime occurs beyond the glass transition temperature by definition, and the melting of feedstock constituents is critical to pelleting without artificial binders.

Compressibility can also be evaluated at lower pressures. At lower pressures (~100 kPa), materials are still in the elastic range. Little particle deformation takes place. Rather, changes in bulk density, measured as volumetric strain, are due to the rearrangement of particles. One apparatus for the evaluation of compressibility is the cubical triaxial tester.

2.3.6 Cubical Triaxial Tester

A cubical triaxial tester (CTT) (Li and Puri, 2003) is commonly used in the powder mechanics field to evaluate the volumetric strain and bulk moduli of powders. Volumetric strain is a measure of the change in bulk volume under an applied load. The CTT uniformly loads a predetermined volume of the bulk material by applying isotropic pressure to a rubber membrane that surrounds the loading cell. The loading schematic for the CTT is presented as Figure 2-3.

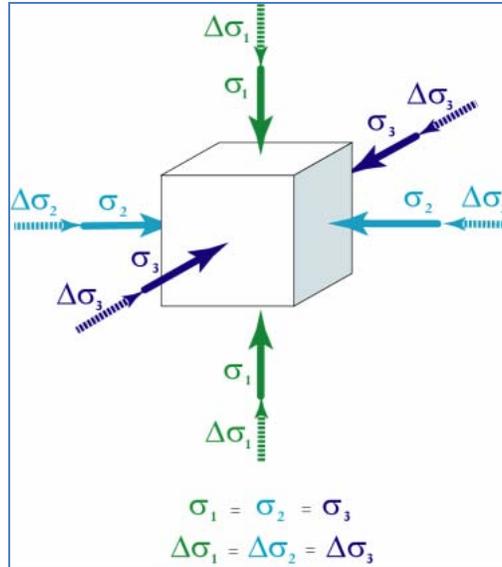


Figure 2-2 – Pressure loading schematic for cubical triaxial tester (Yi et al., 2008)

The cell is repeatedly loaded and relaxed by increasing and decreasing pressure on the cell. Volumetric strain is determined by six linear motion potentiometers configured in the same manner as the forces displayed in Figure 2-2. Data are typically presented as a plot of volumetric strain vs. pressure similar to Figure 2-3.

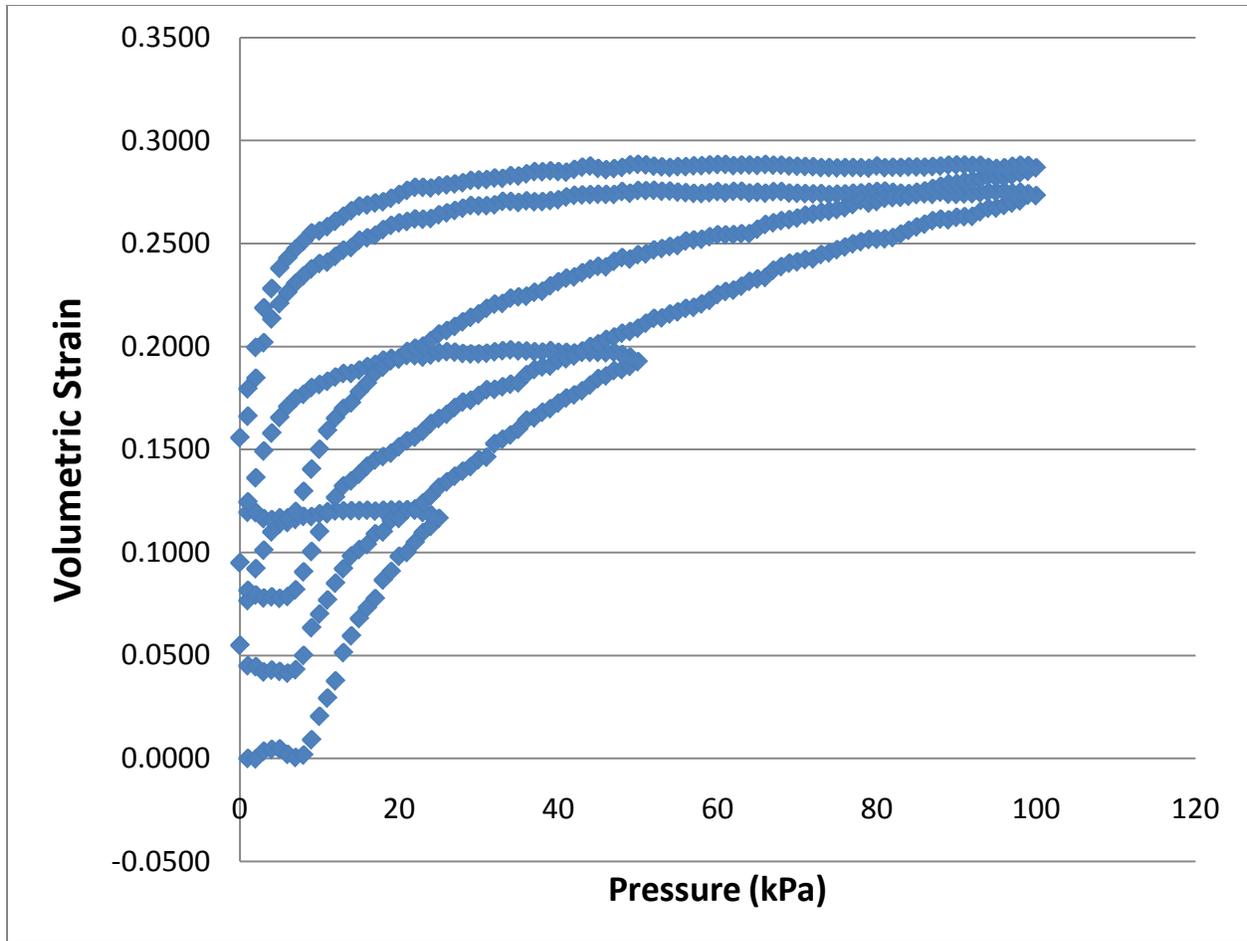


Figure 2-3 – Typical presentation of CTT results

From the plot above, bulk modulus and elastic response are calculated. Bulk modulus is the slope of the curve as the material is being unloaded and then reloaded (Figure 2-4). Bulk modulus is an indicator of the hardness of the bulk material under load. Elastic response is an indication of the material’s tendency to “spring back” when the load is released.

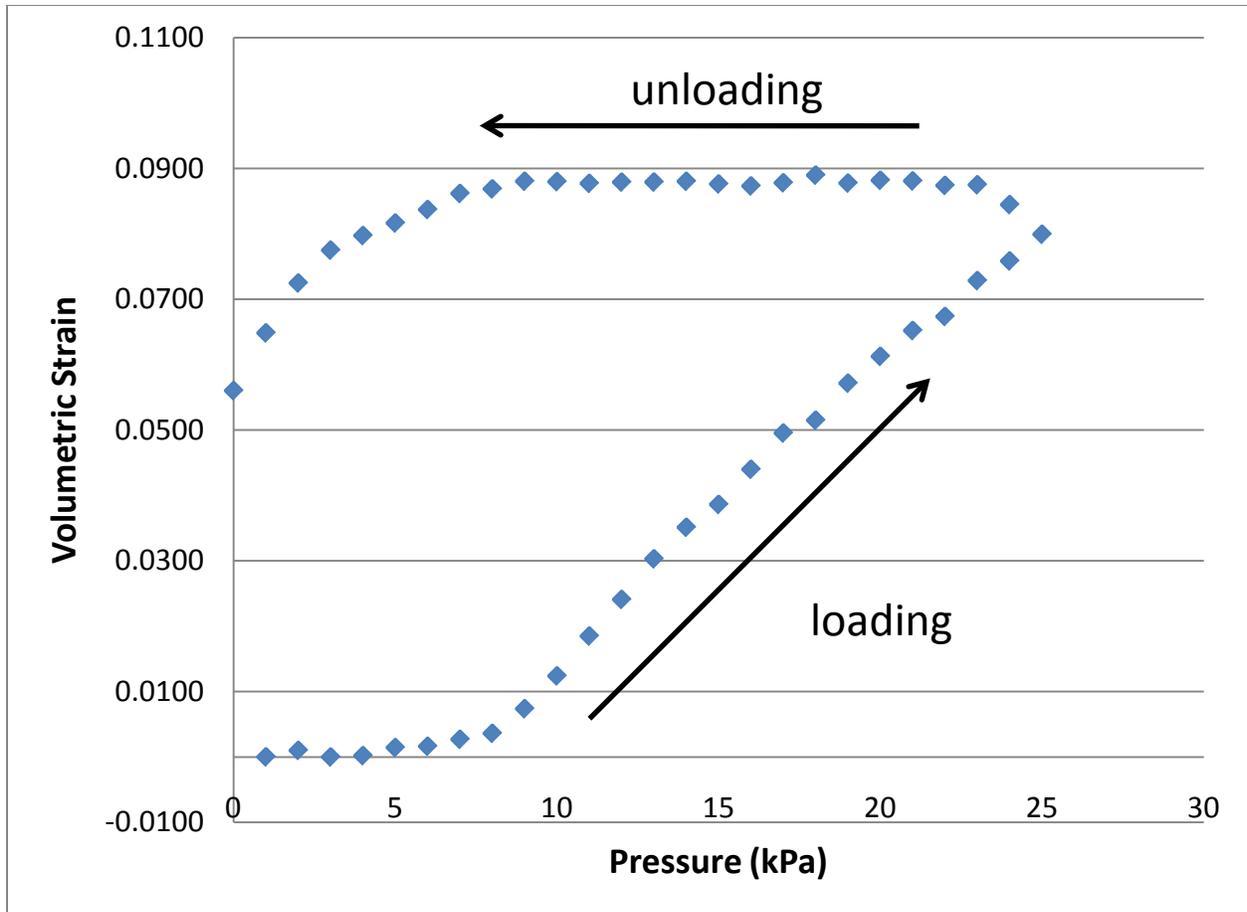


Figure 2-4 – Loading and unloading curves in CTT results

Bulk modulus is calculated for each predetermined pressure preset. In Figure 2-3, pressure presets are 25, 50, and 100 kPa. For simplicity, bulk modulus is taken to be the average slope of the inverse of the unloading and reloading curves as the sample is unloaded and then reloaded to the previous pressure preset. An approximate bulk modulus is shown in Figure 2-5 to illustrate this concept. The figure displays the bulk modulus at 25 kPa.

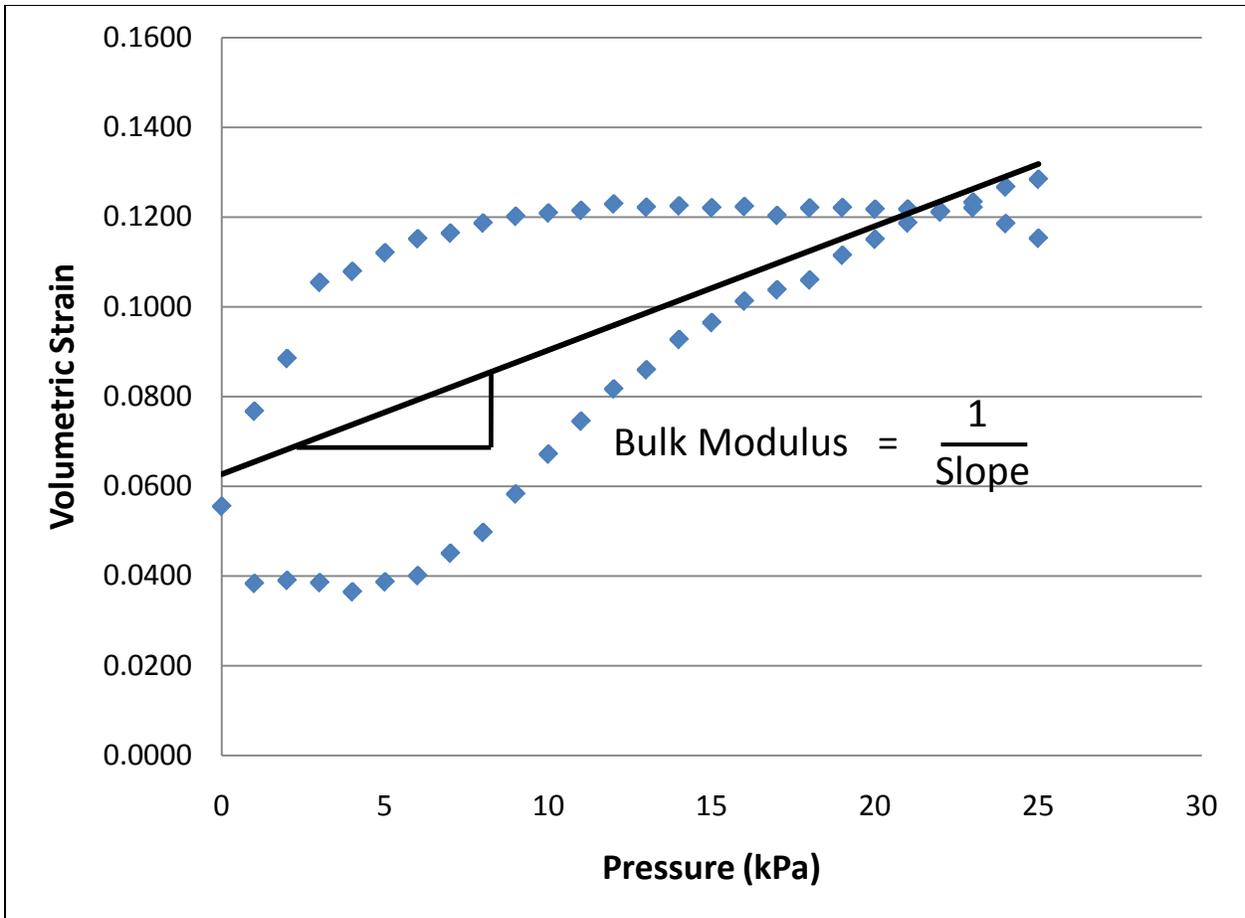


Figure 2-5 – Bulk Modulus

In addition, by comparing volumetric strain values when the sample is loaded to those after unloading, the elastic response of the material is observed (Figure 2-6).

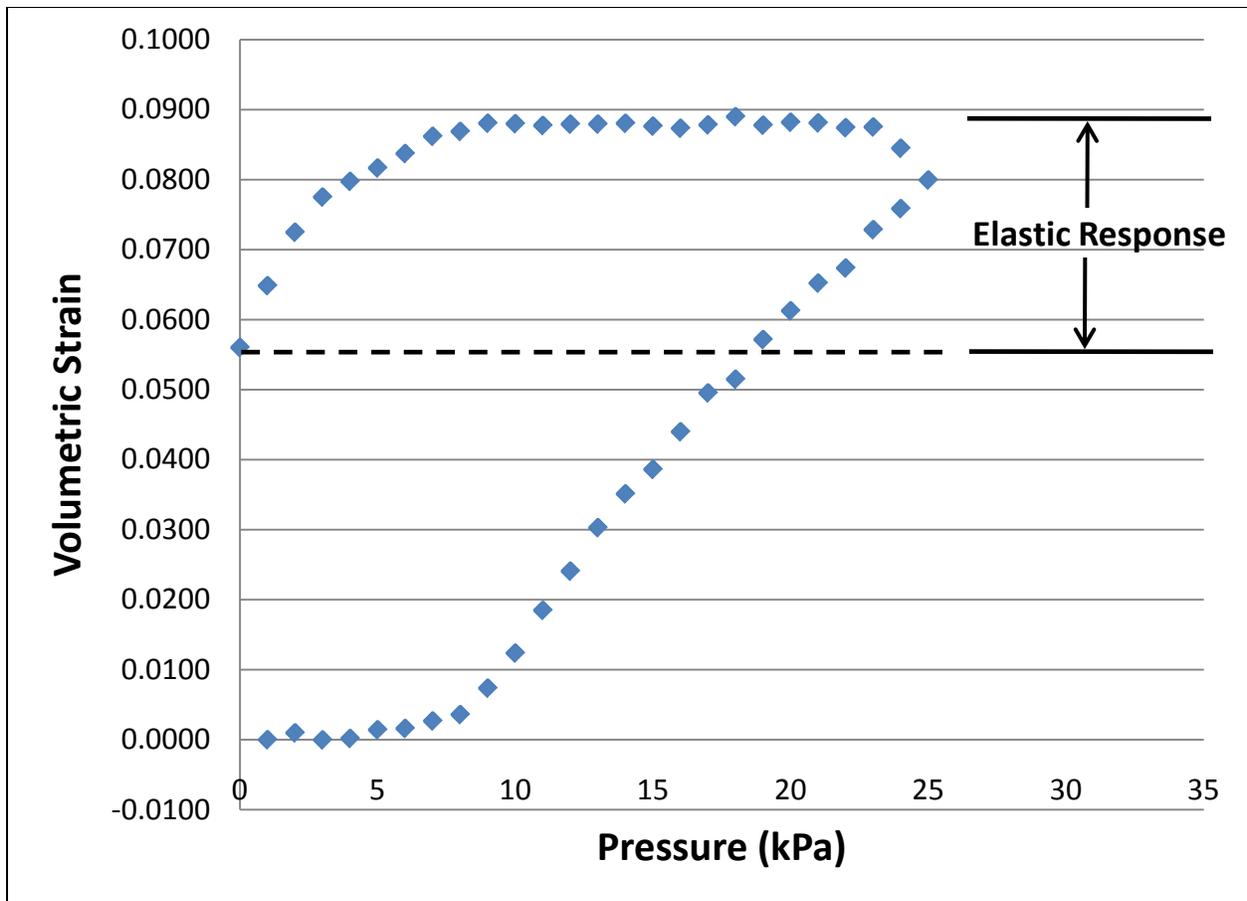


Figure 2-6 – Elastic response from CTT output

By observing the difference between volumetric strain values when the sample is loaded and unloaded, some indication of the material's elastic response can be observed. The greater this difference, the more elastic the material.

2.3.7 Particle Characteristics

Cubical triaxial testing and similar bulk material methods can serve to shed light on the compaction behavior of bulk materials. While bulk material response is ultimately of interest, the behavior of the bulk material is driven by individual particle interactions. In order to explain the behavior of bulk materials, it can be helpful to characterize the individual particles within.

The effect of particle shape and surface roughness on the compaction behavior of bulk materials is well documented within the powder technology and engineered wood fields, but has received relatively minimal attention in the biomass densification literature.

In their work with pharmaceutical powders, Li and Puri (2003) note that particle density, shape, strength and surface characteristics are important in determining the load-response of a system. Similarly, Miyamoto et al. (2002) observe that the thickness swelling, internal bond strength, and linear expansion of particle board are all impacted by particle shape. Like all characteristics of irregular particle distributions, parameters such as particle shape and surface roughness can be difficult to measure, and efforts to quantify them result in large amounts of uncertainty. However, examining these characteristics, even qualitatively, can provide some information regarding compaction behavior.

2.3.8 Lignin Evaluation Method

The bulk of the work in the densification literature evaluates lignin content of feedstocks based on the acid detergent lignin (ADL) method originally developed for the evaluation of livestock feed rations (Shaw, 2008; Mani et al., 2006). The pulp and paper industry, which relies heavily on thermochemical reactions and thermoplastic behavior in production technologies, employs the Klason lignin method for the determination of lignin content.

The ADL method consists of repeatedly washing the sample in detergents of increasingly lower pH, and evaluating fiber constituents based on a mass balance. The sample is first washed in a neutral detergent to evaluate neutral detergent fiber. The sample is then washed in an acid detergent to evaluate acid detergent fiber before finally being washed with strong sulfuric acid for the evaluation of acid detergent lignin. At each step, fiber content is taken to be the

difference in mass by percent from the previous step. By measuring lignin content last, any error in the measurement is compounded. It should be noted that the purpose of the ADL method is primarily to evaluate crude fiber and protein for feed rations, and not expressly to determine lignin content.

By contrast, the evaluation of Klason lignin involves only one acid wash, after which lignin is evaluated by mass. As such, the Klason method is a more direct evaluation of lignin content; Klason lignin evaluation consistently results in substantially higher values of lignin content than the ADL method. Table 2-1 below displays the values of ADL and Klason lignin determined by Joachim et al. (1999).

Table 2-1 – Lignin composition by method

sample	Method	
	ADL	Klason lignin
alfalfa	7.1	15.0
alfalfa	6.6	13.6
Kura clover	2.9	8.4
annual medic	3.6	9.3
maize silage	2.4	12.2
maize silage	2.5	10.0
orchardgrass	1.9	12.6
smooth bromegrass	4.2	15.5
switchgrass	3.0	14.2
oat straw	4.6	16.6

While there is a positive relationship between the two methods, the absolute level of lignin found using the Klason method is much higher than that of the ADL method. Previous researchers had attributed this difference to overestimation by the Klason method (Van Soest, 1967, Lai and Sarkanani, 1971). This claim was, however, dispelled by Jung et al. (1999) via bomb calorimetry. Jung and his colleagues cleverly asserted that, since all other components between the two

methods are the same, the Klason lignin method should overestimate total energy content of the samples. By comparing the measured forage gross energy value to that of the computational analysis of the samples, they find that the ADL method only accounted for 68-84% of the measured gross energy whereas the Klason lignin method accounted for 85-97% of the gross energy (Jung et al. 1999). Thus the ADL method underestimates true lignin content, and Klason lignin is the more accurate method. When examining the effect of lignin content on the densification of biomass, i.e. where thermoplastic behavior is a critical component, it is important to use the method designed to capture this type of behavior.

2.3.8 Process Variables

The parameters addressed in the previous three sections are those of the feedstock entering the pelleting process. Process variables refer to those parameters that are inherent to the pellet mill itself, specifically die dimensions, die speed and the gap between the roller and die.

Industrial pellet dies consist of an annular matrix of perforations characterized by length to diameter ratios (L/D). Length refers to the depth of the die itself and diameter refers to that of the perforations. Generally, pellet durability increases with L/D ratio due to increased shear forces resulting from increased friction between feedstock and die. However, too large a ratio will block the die and choke the mill. This was phenomenon was originally observed by Heffner and Pfof (1973). It is important to note that softwood pellet producers commonly use a deeper die than hardwood producers.

Die speed refers to the tangential velocity of the rollers during the pelleting process. Thomas et al. (1997) observed that high die speeds (10 m/s) were appropriate for small pellets (3 to 6 mm diameter) and lower die speeds should be used in the formation of larger pellets (6 to 7 mm

diameter). Additionally, Heinemans (1991) noted that materials with low bulk densities should be pelleted at low die speeds (4-5 m/s) due to the significant amount of air that must be expelled before desired density is achieved.

The gap between the roller and die refers to the space between the annular matrix and the roller that forces the feedstock through the die. Robohm and Apelt (1989) found that the optimal gap for producing the most durable pig feed pellets was between 2.0 and 2.5 mm. Further increasing the gap to 4.0 to 5.0 mm significantly reduced pellet hardness and durability.

2.4 Quantifying Pellet Strength and Durability

In the investigation of the characterization of a quality pellet, it was evident that a number of definitions exists regarding the strength and durability of pellets. Kaliyan and Morey (2006) present clear definitions of each. Strength and durability in this paper refer to the definitions established in their work and are enumerated in the subsequent sections.

2.4.1 Strength

Strength refers to both the compressive and impact resistance of a pellet. Compressive resistance testing simulates the loading due to self-weight in storage and the crushing of pellets in a screw conveyor. Impact resistance testing models the impact forces induced on pellets during handling in the filling of silos, bins or storage bays when pellets are dropped either on a hard floor or onto one another. Strength is most important when considering bulk delivery systems, which are uncommon in the United States.

Compressive resistance, also referred to as hardness, is defined as the maximum compressive load that a pellet can incur before cracking. Compressive resistance is modeled using a diametrical compression test in which a single pellet is placed between two flat, parallel platens

and an increasing load is applied at a constant rate until fracture. The load at fracture is read off of a recorded stress-strain curve and referred to as the compressive strength of the pellet (Kaliyan and Morey, 2006).

Several methods have been used to establish the impact resistance of densified materials. All involve dropping a single particle several times from an established height and recording the mass or number of pieces retained above a specified particle size. ASTM method D440-86 (ASTM, 1998) of a drop-shatter test for coal was employed by Li and Liu (2000) for testing the durability of biomass logs. An impact resistance index (IRI) (Richards, 1990) was then calculated using equation 1.

$$IRI = 100 \left(\frac{N}{n} \right) \quad (1)$$

where

N = number of drops

n = total number of pieces after N drops

Since the standard number of drops employed by Li and Liu (2000) was always two, the maximum value of IRI was 200. Since briquettes often broke into many small pieces, particles weighing less than 5% of the original mass of the log were not considered in n and were not included in the second drop.

Sah et al. (1980), Khankari et al. (1989), Shrivastava et al. (1989), and Al-Widyan and Al-Jalil (2001) used an impact resistance test to determine the durability of biomass pellets and briquettes.

Pellets were dropped from a height of 1.85 m onto a metal plate four times. Impact resistance was defined as the percentage of the initial weight retained after dropping. Lindley and Vossoughi (1989) employed a similar method to measure the impact resistance of briquettes. Individual briquettes were dropped 10 times from a height of 1.0 m onto a concrete floor and percent loss was calculated.

2.4.2 Durability

Durability is defined by Kaliyan and Morey (2006) as the abrasive resistance of a densified product. Durability, as defined herein, is the most prevalent form of pellet quality analysis employed by pellet manufacturers and is used to adjust parameters during the pelleting process (Winowiski, 1998).

There are two distinct classifications of durability tests. Mechanical tests simulate the forces experienced by pellets in screw conveyance (augers), and model the handling systems commonly employed in the United States. In Europe, handling is largely conducted with the use of pneumatics. Pneumatic tests were developed to simulate the impacts experienced in pneumatic conveyance. The handling methods of the product during transportation and storage should determine which test is used in the analysis of pellet durability. The Holmen test (Franke and Ray, 2006) and Ligno test (Winowiski, 1998) are commonly used to simulate the forces induced on pellets during pneumatic conveyance and will not be addressed at length here. The tumbling can method (ASABE Standards, 2003) is the most common method used by feed manufacturers in the United States (Winowiski, 1998). The Dural Tester was developed at the Agricultural Process Engineering Laboratory at the University of Saskatchewan and has distinct advantages over the tumbling can method (Sokhansanj and Crerar, 2006).

The equipment design and standard methodology for the tumbling can tester are defined by ASABE Standard S269.4 (ASABE, 2003). The tester itself consist of a rectangular box with inside dimensions 30.5 x 30.5 x 12.7cm with a 5.1 x 22.9 cm baffle fixed inside the box on a diagonal of one of the square sides. The box must be sealed so that no dust can escape during tumbling. The tumbling can method calls for a 500 g sample of pellets to be tumbled for 10 minutes at 50 rpm. Following tumbling, the sample is sieved using a sieve size approximately 0.8 times that of the pellet diameter. It is not necessarily the absolute mesh size that matters but the fact that the same mesh size is used before and after tumbling. By using the same screen size, a consistent definition for fines is established, and results will be repeatable. The ASABE Standard (S269.4) calls for two replications of the test. Pellet durability index, or PDI, is expressed as percentage of the initial mass retained on the sieve and is calculated using equation 2.

$$PDI = 100 \left(\frac{M_f}{M_i} \right) \quad (2)$$

where:

PDI = Percent durability index

M_i = Initial mass of pellets

M_f = Final mass after tumbling

The tumbling can method has been modified by several mill operators and researchers with the addition of ball bearings or hex nuts to increase the rigor of the test and obtain a wider range of values (Kaliyan and Morey, 2006).

The Dural tester (Sokhansanj and Crerar, 2006) is similar in design to a food blender. It contains an impeller with blades at a 45 degree angle inside of a canister. The method for sample preparation calls for a 100 g sample to be hand sieved 30 times through a sieve 0.9 times that of the pellet diameter. As with the tumbling can method, achieving a sieve size exactly 90% of pellet diameter is unlikely; again, however, consistency is more important than accuracy.

Following sieving, the sample is placed in the tester, which is run at 1600 rpm for 30 seconds. The sample is again sieved (30 times) by hand and durability calculated as a percentage of the initial weight retained on the sieve after testing using equation 3.

$$D = 100 \left(\frac{M_f}{M_i} \right) \quad (3)$$

where:

D = Durability (%)

M_i = Initial mass of pellets after sieving

M_f = Final mass after sieving

The Dural tester holds several distinct advantages over the tumbling can method. First, it is more rigorous; it yields a wider range of values that are more realistic than those from the tumbling

can method. Hill and Pulkinen (1998) estimated that breakage in pellet shipments was approximately 30% after transport. The ASABE tumbler method typically yields PDI values between 80 and 100. The Dural tester provides a more accurate representation of durability values, yielding values ranging from 5 to 90% for pellets that were predicted by the tumbling can method to have durabilities between 85 and 95% (Sokhansanj and Crerar, 2006). Other marked advantages of the Dural tester include the requirement of a smaller sample and shorter testing duration (30 s vs. 15 to 20 min) than the tumbling can method. The shorter duration is a distinct advantage for quality control, as operators can more quickly establish pellet durability values and adjust parameters accordingly. While the Dural tester is clearly a more effective and robust method for evaluating pellet durability, it has not been adopted by the pellet industry. The standard in the pellet industry remains the tumbling can method. Further, of the quality metrics, durability is the best indicator of pellets' ability to withstand the transportation, storage and handling processes that is considered by the pellet industry.

2.5 Post Production Conditions

Post production conditions play a significant role in both strength and durability values. The timing of measurement in addition to cooling and drying conditions are identified by Kaliyan and Morey (2006) to impact durability and strength. Shocks in moisture content during storage and handling can reduce the structural integrity of pellets, and should be avoided. Moisture content considerations, including shocks in relative humidity and exposure to the elements, are similar to those of grains and other hygroscopic commodities and pellets should be handled with the same considerations in mind.

2.5.1 Timing of Measurement

Kaliyan and Morey (2006) distinguish between the “green” strength and the cured strength of pellets. Green strength refers to measurement carried out immediately after production and cured strength refers to testing conducted approximately one week later. Timing of measurement affects both durability and compressive resistance measurements.

In their work concerning the durability of wood, alfalfa, and hay wafers, Mohsenin and Zaske (1976) observed that the timing of abrasion testing was indirectly related to durability values. The durability of the densified products tested immediately after production was higher than those obtained 45 minutes later. This was attributed to the negative impact of drying and expansion on pellet durability.

Payne (1978) observed that the timing of measurement significantly impacted the hardness values of dairy feed pellets when measured immediately after production and 24 hours later. Compressive resistance increased from 78.5 to 131.4 N with additional curing. He attributed this to the formation of solid bridges as the pellets cooled.

2.5.2 Cooling and Drying

Due to friction experienced in pelleting, pellets exit the process at temperatures and moisture contents higher than that of ambient equilibrium. Immediately after production, pellets are cooled and dried, usually with the use of forced air, to within 5 degrees C and 0.5% of equilibrium moisture content at ambient conditions (Turner, 1995). Cooling and moisture reduction that occurs too rapidly can result in the cracking of pellets. Proper cooling promotes the formation of solid bridges and increases the viscosity of liquid components, improving the overall structural integrity of pellets (Thomas et al., 1997).

2.6 Development of Acceptable Levels of Strength and Durability

The establishment of criteria for suitable levels of strength and durability is desirable to ensure a uniform product in the interest of maintaining consistent quality. Consistent quality is essential to ensure acceptance in the marketplace. Criteria regarding the strength and durability of pellets should consider the rigors of handling, transportation, and storage and weather conditions of the locations where the products are transported/exported (Khoshtaghaza et al., 1999). Weather conditions will not be considered herein, as suitable environmental conditions should be provided by the handler similar to the handling of grains and other hygroscopic commodities. The United States Pellet Fuel Institute (PFI) recently adopted new quality standards that include durability requirements. The standards are included as Appendix A. The measurement of durability is essential to characterizing pellet quality, since it predicts pellet performance in transportation, storage, and handling.

Chapter 3

Hypothesis and Objectives

Densifying biomass serves to increase the feedstock bulk density for more efficient transport, storage, and handling. Pellets are the most widely produced form of densified biomass, and are generally utilized in pellet stoves for residential heating. Pellet durability is largely a function of feedstock and process parameters. In examining the literature, the only variable expected to change with tree species is lignin content.

It is proposed here that the effect of tree species on pellet durability is through variation in lignin content. The null and alternative hypotheses to be tested are:

H_0 = Variation in lignin content cannot explain differences in durability values between pellets produced from different tree species.

H_a = Variation in lignin content can explain differences in durability values between pellets produced from different tree species.

The goal of this research effort is to test the above hypothesis. The specific objectives in support of this goal are:

1. Evaluate the lignin content of the various tree species using the Klason method of lignin determination
2. Produce pellets using two distinct treatments:
 - a. pure species displaying a range of lignin values
 - b. mixed species across the same range of lignin values
3. Evaluate pellet durability of each
4. Compare durability results between treatments to determine if lignin content alone predicts variation between species

Pellets of each species and species mix are produced over a range of moisture contents in order to ensure that any interactive effect between lignin content and moisture is captured. Given the relationship between moisture content and the glass transition temperature of lignin, an interactive effect is likely.

By isolating the effect of tree species, the goal is to better predict compaction behavior without putting material through the die. The ability to predict pelleting performance of a feedstock without putting it through the mill can improve productivity by avoiding the plugging of dies. Plugged dies inevitably lead to mill downtime and repair costs, as the replacement of a plugged

die is no small task and the die is an expensive part of any pellet mill. Plugged dies are often rendered useless, as efforts to drill the plugged material from the die tend to foul die holes.

Chapter 4

Methodology

The experimental plan for the evaluation of durability values of biomass pellets and the corresponding breakage levels in storage and handling is discussed in this chapter. After providing a brief overview, each of the three phases is discussed in detail. The three phases of the research plan are:

1. Material characterization and preparation
 - a. Material characterization
 - i. Lignin content
 - ii. Particle size distribution
 - iii. Bulk modulus and volumetric strain
 - iv. Particle shape
 - b. Sample preparation
 - i. Particle size reduction
 - ii. Moisture addition
2. Pellet production
3. Durability evaluation

Material characterization consists of compositional analysis, particle size analysis, triaxial compression testing and evaluating particle shape under a microscope. Material preparation is conducted by first reducing particle size with the same hammermill and screen size, followed by

the addition of moisture. Durability is evaluated via the tumbling can method. Once pellets are produced and durability evaluated, results are analyzed to determine the impacts of the observed and controlled parameters on pellet durability.

4.1 Overview

Wood fiber was obtained from pellet producers and local sawmills. Through industry contacts, samples of pure or well-defined species were obtained from pellet producers across the country. Having obtained and characterized the samples, sample preparation and pellet production commenced. Once pellets were produced, durability was evaluated. Durability results were analyzed to determine whether a relationship existed between lignin content and durability. Having observed no statistically discernable relationship, the effects of bulk modulus and particle shape were investigated as possible causes for variation in pellet quality between species.

4.2 Experimental Phases

The subsequent sections address each phase of the research in detail. By examining the compaction behavior of sawdust from different tree species, parameters affecting the compaction behavior of biomass that might otherwise be overlooked can be isolated.

4.2.1 Material Characterization

Material characterization consists of the evaluation of lignin content, particle size distribution, triaxial compression testing, and particle shape. Since the focus of the research is to determine whether lignin content explains variation between species, Klason lignin of each fiber feedstock was evaluated prior to preparing samples.

Lignin content is evaluated via NREL-CAT Std. No. 001-003 (NREL 2007a). The standard develops methods for characterizing cellulose, hemicelluloses, and lignin content among other chemical constituents.

First, the sample is milled in a Wiley mill using a 2 mm screen. Next, the method calls for a 0.3 g (+/- 0.01 g) sample of fiber to be massed and placed into autoclavable glass tubes along with 5 mL of 72% sulfuric acid. After acid addition, samples are placed into a 30°C water bath for 1 hour while swirling each tube once every ten minutes. Following heating, samples are removed from the water bath and the solution diluted to 4% H₂SO₄ and autoclaved for one hour. Once the autoclave cycle is finished, samples are removed and filtered into 500 mL sidearm Erlenmeyer flasks. Samples are filtered using a filtering crucible. Following filtering, the material left in the crucible is dried overnight and is taken to be the amount of Klason lignin in the sample on a mass basis.

NREL recommends that both water and ethanol extractives values be analyzed to accurately determine Klason lignin content. Extractives were analyzed for the pure species, but not for the second treatment, where a red and white oak sample was mixed with pine. While accounting for extractives did affect the lignin content of pure species differently, it was not enough to change the order of species from a lignin content perspective. Given the lack of a trend between lignin and durability in the pure species and the relatively uniform effect of accounting for extractives, subsequent Klason lignin values presented are actually Klason lignin plus extractives values.

Particle size distribution is evaluated following ASABE Standard 319.3. The method was adapted slightly based on the number of sieves that could be placed in the available shaker.

Sieve size selected were US standard numbers 4, 6, 8, 16, 30, 40, 60, 100, and 200 in addition to

finer. Analysis of particle size is presented as both histograms and in terms of calculated mean geometric diameter and geometric standard deviations from the mean using equations 4 and 5, respectively.

$$d_{gw} = \log^{-1} \left[\frac{\sum_{i=1}^n (w_i \log(d_i))}{\sum_{i=1}^n w_i} \right] \quad (4)$$

$$s_{log} = \left[\frac{\sum_{i=1}^n w_i (\log(d_i) - \log(d_{gw}))^2}{\sum_{i=1}^n w_i} \right]^{1/2} \quad (5)$$

where:

d_{gw} – geometric mean diameter

s_{log} – geometric standard deviation

w_i - mass on sieve i

d_i - nominal aperture size of sieve i

n – number of sieves + 1 (pan)

Given the irregular shape of sawdust particles, histograms provide the most accurate characterization of particle size distribution; however, means and standard deviations are calculated for statistical analysis.

Triaxial compression testing consists of placing a sample into a membrane of known volume and repeatedly compressing and relaxing the membrane at increasingly higher pressures. From the triaxial compression test, volumetric strain is measured at each pressure increment as the cell is

loaded. Bulk modulus (K) is then calculated at each pressure set point as the ratio of the pressure to the volumetric strain (4).

$$K = -\frac{P}{\delta V/V} \quad (6)$$

where:

K – Bulk Modulus (kPa)

P - Pressure (kPa)

$\delta V/V$ - volumetric strain (dimensionless)

Bulk modulus can be thought of as the concept of the modulus of elasticity of solid materials applied to bulk solids; like the modulus of elasticity of solids, bulk modulus is an indicator of the hardness of a bulk material. In addition to bulk modulus and volumetric strain, the elastic response of a material can be observed from triaxial compression testing. Elastic response is important in compaction behavior since it is an indicator of the tendency of the bulk material to “spring back” when pressure is released. When pellets exit the die, the elastic response of the material can cause pellets to fracture.

4.2.2 Sample preparation

Sample preparation requires particle size reduction and moisture and lubricant addition. Particle size reduction is required to achieve a consistent, small grind, and moisture addition is necessary to reach the moisture content dictated by experiment design. Lubricant addition was necessary due to the relatively low force achievable by the lab-scale pellet mill available for use.

Particle size reduction is conducted with a C.S. Bell No. 1 Modern Hammermill using a sixteenth inch screen (Figure 4-1).



Figure 4-1 – Picture of C.S. Bell No. 1 Modern hammermill

One 1.59 mm (0.0625 in.) screen was determined by preliminary pellet production to be the optimal screen size of those available for pellet production. Larger screen sizes did not allow material to flow through the die. The same hammermill and screen size were used for all samples in order to achieve the most consistent die grind possible.

Moisture content was measured according to ASABE Standard S358.2. Approximately 10 g of fiber was massed using a balance with accuracy to 0.01 g. Moisture was then added by mass to achieve the desired moisture content. Each sample was massed to 2.26 kg (5 lb.). Three moisture contents (10, 12, and 14% w.b.) were selected for investigation based on preliminary

production. A range of moisture contents was selected due to the expected interactive effect between moisture and lignin. If the level of lignin content does indeed impact pellet durability, it would be expected that different levels of moisture would impact each specie differently.

Following moisture addition, samples were mixed using a mortar mixer and electric drill.

Mixing was necessary to equilibrate moisture content throughout the sample. Further, preliminary production indicated that the timing of moisture addition was important. This is thought to be related to whether the moisture was adsorbed or absorbed by the fiber or simply surface moisture. Therefore, moisture was added 12 hours prior to pellet production. Following moisture addition, samples were sealed so that moisture was not lost to the ambient air during the equilibration period.

During preliminary production, it became evident that lubricant addition was necessary to prevent the die from “choking.” Vegetable oil is commonly used as a die lubricant by hardwood pellet producers to reduce friction in the die. Canola oil was used for this experiment. Based on an iterative process, 9 mL/kg of material was added to each sample.

4.2.3 Pellet production

Pellet production is conducted using a lab-scale pellet mill developed by Buskirk Engineering for Ernst Conservation Seeds (Meadville, PA) (Figure 4-2).



Figure 4-2 – Buskirk Engineering Pellet Mill

The mill has a flat, horizontally configured die. By contrast, more sophisticated lab-scale units and industrial pellet mills have a cylindrical, vertically configured die. In both cases, rollers compress material through perforations in the die by rotating on the die face. However, in the vertical configuration, the die itself rotates, whereas the roller carriage of the horizontal configuration rotates.

Die speed (in this case, roller speed) was set at 50 RPM. Initially, no gap existed between the roller and die; however, the roller carriage deflected upon loading and caused displacement between the rollers and the die. Deflection of the rollers was not measured. Future efforts using a horizontally configured die should evaluate roller deflection.

4.2.4 Durability Evaluation

Following pellet production, pellet durability was evaluated using the tumbling can tester constructed at Penn State in accordance with ASABE Standard S269.1. A photo of the tester is presented below as Figure 4-3.

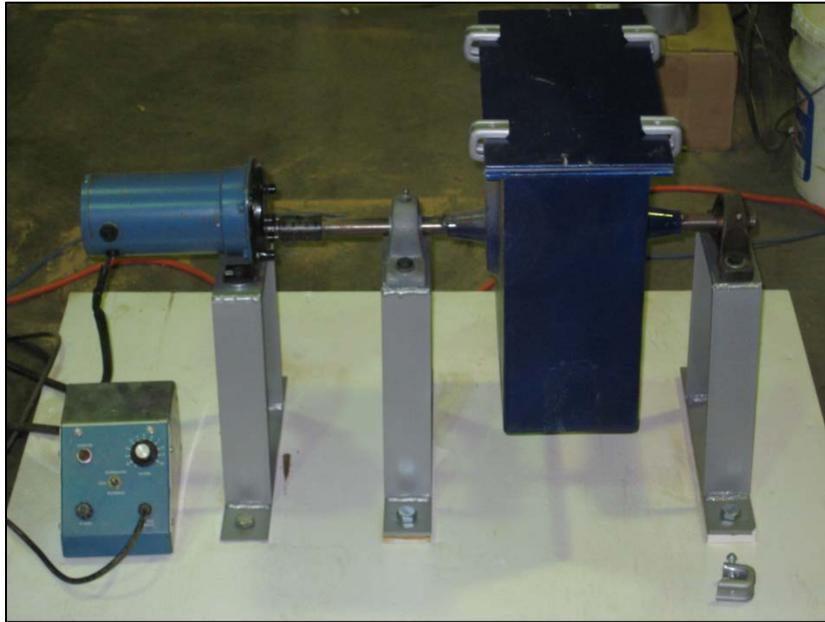


Figure 4-3 – Tumbling can tester

Timing of measurement and storage conditions of pellets were demonstrated in the literature review to impact the evaluation of pellet quality. Following production, pellets were allowed to cool at ambient conditions for approximately one hour before being placed in sealed containers to ensure that they were not exposed to shocks in moisture.

The method for the evaluation of durability using the tumbling can method consists of the following steps:

1. Sieve pellets with sieve size $\sim(0.8)$ pellet diameter
2. Mass 500 g of sample and place in tester
3. Rotate at 50 rpm for 10 minutes
4. Sieve again with same sieve as in step 1
5. Mass material retained on sieve
6. Repeat with new sample

ASABE Standard S269.4 calls for two replications of the above method. The two values are averaged to obtain PDI. Given the relatively small amount of pure species available, the method was adapted by reducing sample size to 250 g. While this does deviate from the Standard, the smaller sample size was maintained throughout the experiment. Further, since relative differences in durability are of interest, the smaller sample size should not affect results. A sieve size of 4.75 cm (no. 4 US standard classification) was used. Again, this is somewhat smaller than the 5.08 cm screen called for by the method, but consistency is what is important.

Chapter 5

Presentation of Results

The presentation of results begins with the characterization of feedstock properties, followed by the evaluation of pellet durability, and concludes with a discussion of bulk material and particle properties that may explain observed differences in durability.

5.1 Material characterization

Material characterization begins with the evaluation of lignin content for the seven different species and species mixes from which samples were prepared. Next, particle size analysis results are presented by displaying a typical histogram of size distribution and reporting the geometric

mean diameters and geometric standard deviations of each feedstock. After characterizing particle size distribution, triaxial compression testing results are presented for selected species.

5.1.1 Lignin content

The NREL method calls for the evaluation of extractives content to determine the true level of Klason lignin. However, when Klason lignin was first analyzed, the BioWaste Lab in Agricultural and Biological Engineering did not have the capability to analyze water and ethanol extractives. Therefore, the values used for the purposes of this experiment are Klason lignin plus extractives. Klason lignin plus extractives values ranged between 25.05% for red oak to 30.20% for the cedar/douglas fir mix. Klason lignin plus extractives values are presented for each of the feedstocks in Table 5-1.

Table 5-1 –Klason lignin plus extractives values

species	Klason lignin + extractives (%)	standard deviation	# of observations
pure species			
maple	25.05	1.62	4
red oak	25.77	0.73	6
walnut	26.60	0.03	2
lodgepole pine	28.95	0.47	4
cedar/doug fir	30.17	0.32	2
oak – pine mix			
oak	26.41	0.35	4
pine	30.21	0.86	4

Since the compositional analysis was conducted, the Biowaste Lab has invested in the technology to evaluate ethanol and water extractives. Given the similarity in lignin values, statistical difference between lignin values of pure species was analyzed (Table 5-2).

Table 5-2 – Statistical comparison of lignin values

comparison	pooled deviation	t-stat	df	result
maple - red oak	1.15	0.97	8	fail to reject
maple - walnut	1.41	1.27	4	fail to reject
red oak - walnut	0.30	3.41	6	reject
walnut - lodgepole pine	0.40	6.73	4	reject
lodgepole pine - cedar/dougfir	0.43	3.27	4	reject

Looking at Table 5-2, maple is not statistically different from either red oak or walnut, however, all other species are in fact statistically different from one another. To account for differences in the number of samples between groups, a pooled standard deviation was used for the t-test of difference between means.

To determine whether accounting for extractives would significantly impact results, the five well-defined species were evaluated for extractives content. With the exception of oak and walnut, the order of lignin values did not change for any of the species considered. Extractive-free Klason lignin content is presented in Table 5-3 along with Klason lignin plus extractives values for the sake of comparison.

Table 5-3 – Effect of extractives on Klason lignin content

species	Klason lignin + extractives (%)	Klason lignin (%)
maple	25.0	21.1
red oak	25.8	22.1
walnut	26.6	21.7
lodgepole pine	28.9	25.1
cedar/doug fir	30.2	27.2

Since only one observation is available for Klason lignin plus extractives values³, a statistical test could not be performed. However, the bulk of the error in the Klason lignin evaluation can be attributed to human error in the filtration process, since solids are repeatedly poured into filtering crucibles. This can often lead to solids dripping from the glass tubes down the sides of the crucible. Since Klason lignin is evaluated on a mass basis, this is considered a large source of error. By contrast, the extractives evaluation is an extraction process that calculates the mass of extractives based on infrared chromatography. This method is not as susceptible to human error as the Klason lignin evaluation. Therefore, similar standard deviations are expected in the extractives free values. Based on this observation, it is not anticipated that the small difference in Klason lignin content is significant.

5.1.2 Particle size distribution

Particle size distribution can play a large role in compaction behavior. Both mean particle size and the range of particle sizes present are important factors in pellet formation. Most research regarding particle size distribution indicates that a range of particle sizes is optimal for pellet formation (Payne, 1978; Kaliyan and Morey, 2006a; Shaw, 2008). Fines serve to fill the pore space created by larger particles and promote the development of interparticle bonds. In an effort to control for particle size, the same hammermill and screen size were used for the size reduction of all samples.

Particle size was evaluated both graphically and numerically. Geometric mean diameter (equation 4) and geometric standard deviation (equation 5) for each of the five samples of pure species is presented as Table 5-4.

³ Due to extractives equipment breakdown and time constraints, only one extractives test was able to be run

Table 5-4 – Particle size statistics

sample	mean diameter (mm)	standard deviation (mm)
red oak	0.434	0.343
cedar/doug fir	0.446	0.272
walnut	0.452	0.271
maple	0.452	0.267
lodgepole pine	0.478	0.380

Only one observation of each sample was made; however, given the similarity in mean diameter values and histograms and the relatively high standard deviations, additional replications are not expected to yield a different result. From Table 5-4, it is clear that there is no discernable difference between geometric mean particle diameters for any of the species analyzed. The calculation of geometric mean diameter and standard deviation are predicated on the assumption that the particle size being analyzed displays a lognormal distribution. A histogram of the particle size distribution of the maple sample is presented as Figure 5-1 to illustrate the nature of the distribution.

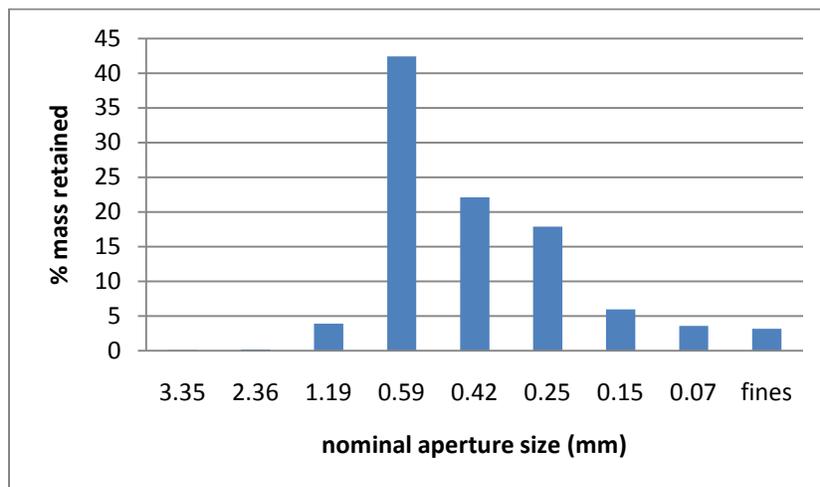


Figure 5-1 – Histogram of maple particle size distribution

Figure 5-1 is typical of the particle size distributions measured. The histogram validates the use of geometric mean diameter and geometric standard deviations; the range of particle sizes is consistent with a lognormal distribution. Particle size distributions of the remainder of feedstocks are presented in Appendix B.

5.1.3 Cubical Triaxial Testing

Triaxial compression testing is often used in the geology and powder technology fields to characterize the compression behavior of bulk materials. The method is applied here to determine the compression and relaxation behavior of the sawdust feedstocks. From the CTT, bulk modulus and volumetric strain are determined.

Another bulk material property that is of interest is the elastic response of the material when the load is released from the sample. By observing the amount of relaxation as the load is released from the sample, some indication of the material's tendency to "spring-back" can be gleaned. This can be detrimental to both pellet formation and the density of the packed bed. The density of the packed bed is important since it is the initial density of the material entering the die. Plots of pressure vs. volumetric strain for red oak and lodgepole pine at several moisture contents are presented in Appendix C.

5.2 Pellet Production

Pellet production was first conducted for each of the pure species. With the exception of the lodgepole pine sample at 10% MC, whole pellet formation was observed for each of the pure species. With the lodgepole pine sample, however, the product took on the shape of small disks. Whole pellets and the disks produced from red oak and lodgepole pine samples at 10% MC are shown in Figure 5-2.



Figure 5-2 – Comparison of Red Oak and Lodgepole Pine Pellets

Looking closely at the whole pellets produced from red oak (Figure 5-3), striations can be seen in the pellet.



Figure 5-3 – Picture of Red Oak Pellets Highlighting Striations

Interestingly, these striations serve as fault points when breaking the pellet along its radial axis. In fact, breaking the red oak pellets apart by hand yields a product nearly identical to that of the lodgepole pine (Figure 5-4).



Figure 5-4 – Picture Showing Disk Structure of Crumbled Pellets

From this observation, the conclusion is drawn that the pellets are comprised of many of these small disks packed together and bonded. The most likely bonding mechanism is solid bridges. The disks are formed before the feedstock is extruded in what is referred to as the “packed bed.” The packed bed is the material between the die and roller that serves to “precompress” the feedstock before extrusion. Likely, the packed bed holds its shape via mechanical interlocking. Each pass of the roller serves to add a “disk” to the die hole. When successful pellets form, it is because these disks are packed closely enough and experience enough heat to reach the glass transition temperature of the inherent binders in the feedstock. At this point, the disks “melt” together to form a pellet. When these bonds are sufficiently strong, material exits the die in the form of a whole pellet. When these bonds are weak or do not form at all, material exits the die in

the form of disks. Interestingly, a loud “popping” can be heard when the material is forming these disks and not whole pellets. It is similar to a popcorn popper in sound, and is thought to be the individual disks relaxing and shooting out of the die. This discussion is continued later in the chapter.

5.2.1 Lignin Content and Durability

Pellets were produced from both pure species and species mixes intended to closely replicate the range of lignin contents found in the pure species. Pure species lignin contents ranged from 25.1 to 30.2 percent, respectively. Mixed species lignin contents did not capture the lower range, but did represent the bulk of lignin contents from the pure species, ranging from 26.5 to 30.2 percent.

For the pure species, pellets were produced for all moisture contents, whereas problems with the pellet mill prevented the mixed species from being produced at 14% MC. However, the primary purpose of pellet production was to investigate the effect of lignin content on pellet production. Plots of durability vs. lignin content for the pure and mixed species treatments are presented as Figures 5-5 and 5-6, respectively.

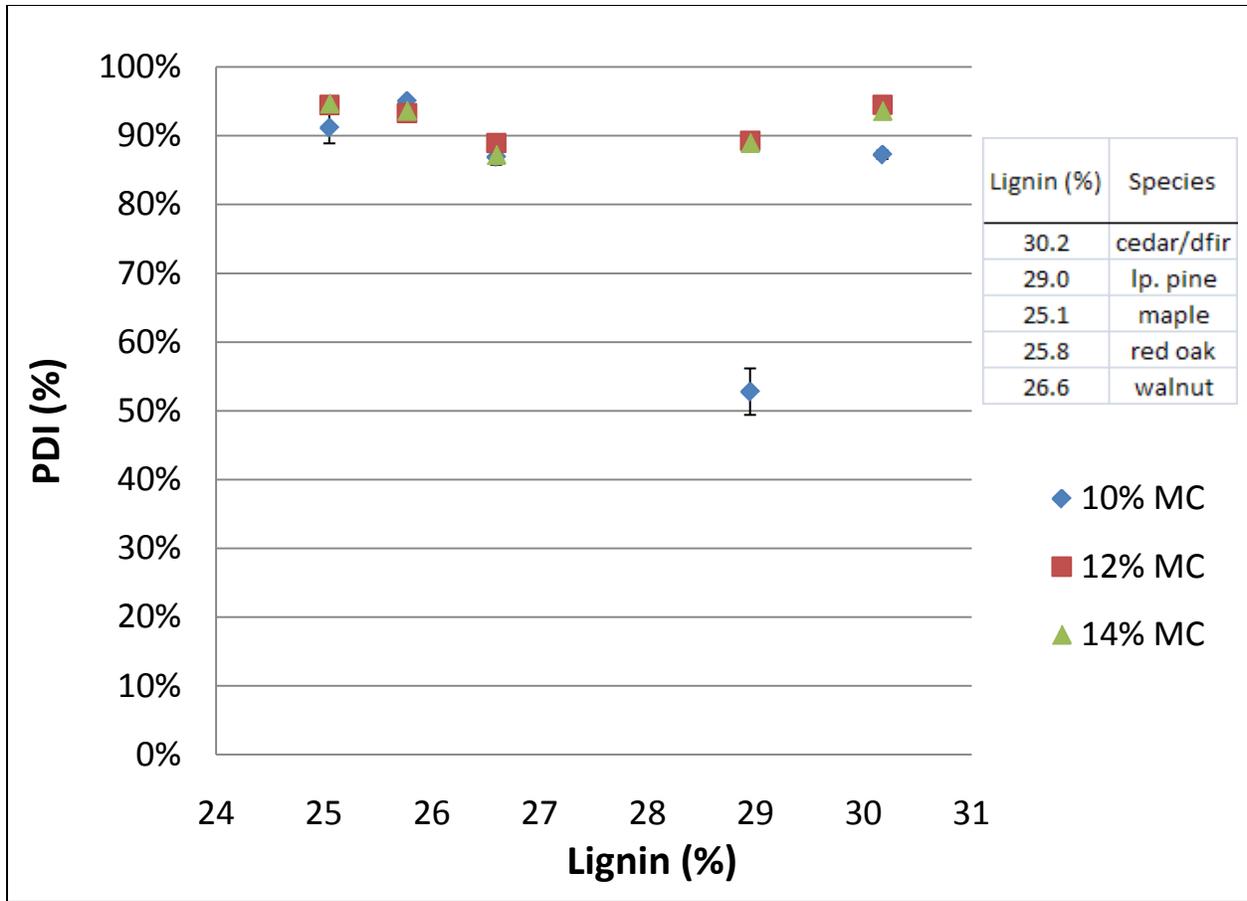


Figure 5-5 – Durability (PDI) vs. Lignin Content for Pure Species

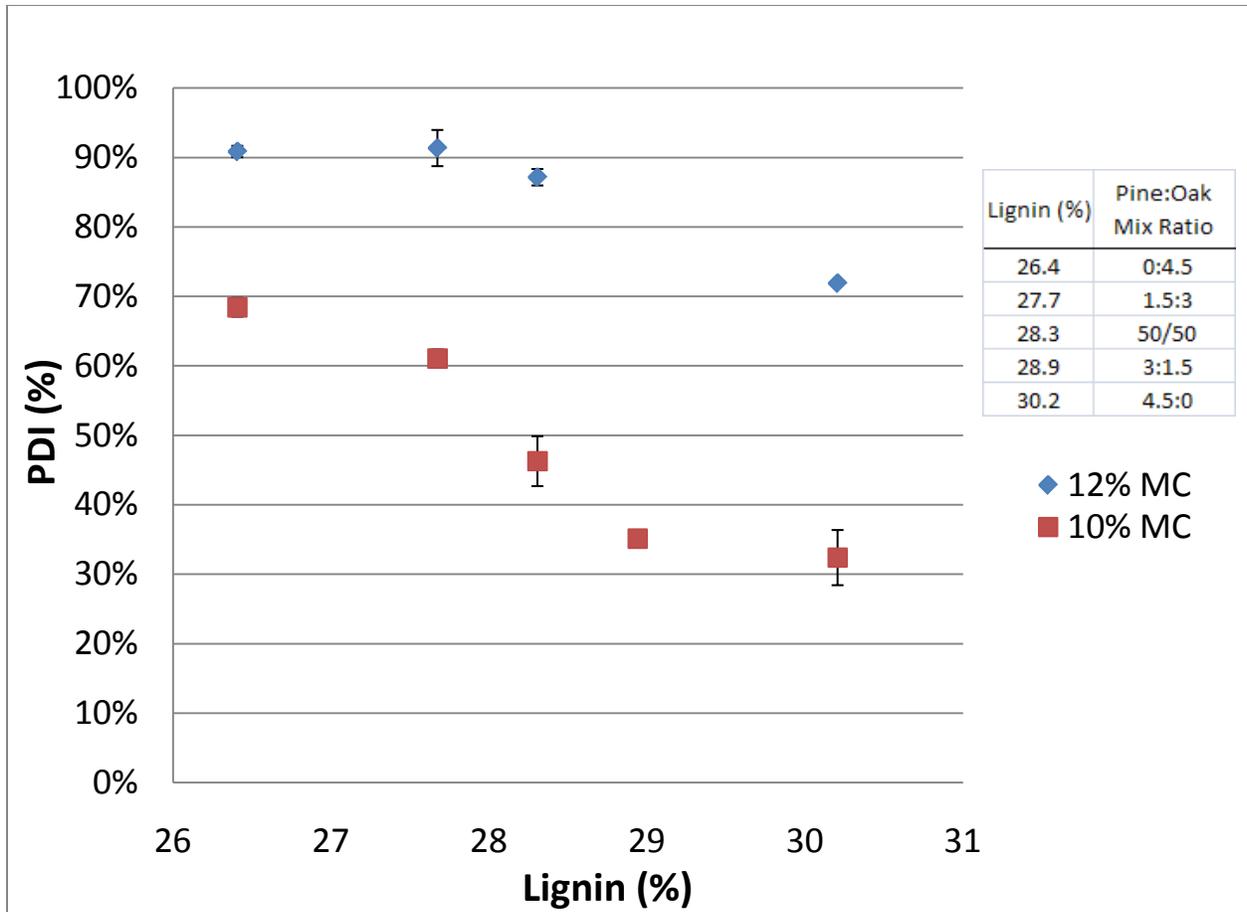


Figure 5-6 – Durability (PDI) vs. Lignin Content for Mixed Species

As can be seen from the figures above, no consistent relationship was found between lignin content and pellet durability at any moisture content. In fact, pellets produced from the mixed species become less durable as lignin content increases.

In nearly all cases, optimal moisture content was 12%. The two exceptions being red oak (lignin = 25.8%) and maple (lignin = 25.05%). For these species, the optimal moisture contents were 10 and 14%, respectively. The durability value for the red oak at 10% MC is thought to be artificially high, since the bulk of this sample was produced with less lubricant.⁴ The durability

⁴ Initially, 15 mL oil/lb sample was added. Red oak was the first sample and displayed excessive heating (black pellets). Subsequent samples were produced with 20 ml oil/lb sample.

of the maple sample at 14% MC (94.5) was not significantly different⁵ from that at 12% (94.7). Lignin content, durability values, and bulk density for each sample produced are presented in Appendix D.

For the mixed species, durability falls off dramatically as lignin content increases. However, given the lack of a relationship between lignin content and durability found with the pure species, this effect cannot be attributed to lignin content. The species mixed were pure pine and a red and white oak mix. Generally, the greater the hardwood content, the higher the durability value. In order to gain insight as to possible reasons for this relationship, differences in bulk material characteristics are investigated between the pure species materials that produced the most and least durable pellets, red oak and lodgepole pine, respectively.

5.2.2 Bulk Material Properties

Having observed that particle sizes are not statistically different, the focus of bulk material properties that may influence the densification process are bulk modulus and elastic response of the feedstock. Bulk modulus is taken to be the inverse of the slope of the volumetric strain vs. pressure curve as pressure is increasing, whereas elastic response is the difference between volumetric strain values when the sample is loaded compared to that when unloaded.

Differences in these parameters for red oak and lodgepole pine are the focus of the bulk material discussion.

In all cases, bulk modulus for the oak sample was much greater than for lodgepole pine (Table 5-5).

⁵ t-val=.0342, n=4, reject

Table 5-5 – Bulk Modulus Comparison

Bulk Modulus (kPa)				
Pressure (kPa)	Lodgepole Pine	Red Oak	t-stat ($\alpha=.05, n=4$)	result
25	178.3	292.0	29.65	reject
50	291.5	429.7	25.06	Reject
100	512.4	678.1	6.84	Reject

This indicates that red oak, in bulk, is “harder” than lodgepole pine in bulk. The likely effect of the greater bulk modulus of oak is to generate greater friction, and thus more heat, while being pelleted. This offers some explanation as to the lack of quality pellets produced from the lodgepole pine sample at low moisture content. The logic behind this explanation is that the amount of friction experienced by the lodgepole pine sample was not sufficient to reach the glass transition temperature of the pine at the relatively low moisture content (10%). However, the higher bulk modulus observed for the oak sample was sufficient to generate enough friction (therefore, heat) for the sample to reach the glass transition temperature, even at the lower moisture content. As moisture was added to the pine, it is postulated that this had the effect of lowering glass transition temperature, creating much more durable pellets at a moisture content of 12%.

Further, a greater elastic response was observed for the pine sample than for oak. Elastic response is important in compaction behavior, since it is a measure of the tendency of the bulk material to “spring back” when unloaded. This may serve to explain the popping sound that was heard as the low-moisture pine sample exited the die. Elastic response at each load is presented for both the oak and lodgepole pine samples as Table 5-6.

Table 5-6 – Elastic Response of Red Oak and Lodgepole Pine

Elastic Response				
Pressure (kPa)	Lodgepole Pine	Red Oak	t-stat ($\alpha=.05, df=2$)	Result
25	0.0606	0.0411	21.53	Reject
50	0.0957	0.0739	9.71	Reject
100	0.1538	0.1032	5.27	Reject

Elastic response is the difference in volumetric strain values between loaded and unloaded observations. Looking at the table, volumetric strain decreased approximately 0.02-0.05 more for lodgepole pine than it did for red oak at each pressure increment. This indicates that the pine sample has a greater tendency to “spring-back” when the load is released. This effect can be detrimental to the pelleting process as the material tries to expand upon leaving the die, resulting in fractured pellets.

Softwood pellet producers generally use a deeper die for pelleting than hardwood producers. The reason for this can be attributed to the lower bulk modulus of softwood species. Since softwoods are softer, more area is required to generate the amount of heat (friction) necessary for plastic deformation. Plastic deformation, in the case of biomass pellets, occurs at the glass transition temperature of the lignin in the biomass. The deeper die used by softwood producers is necessary to generate the same amount of heat that occurs in the shallower, hardwood dies.

5.2.3 Particle Shape and Surface Roughness

In an effort to explain observed differences in the elastic response and bulk moduli, particle shape, surface roughness, and particle density are investigated. Given the irregular shape and fibrous nature of the particles, mechanical interlocking is thought to play a substantial role in the compaction mechanism. In fact, Gray (1968) had observed that mechanical interlocking may

provide enough strength to resist cracking during elastic recovery. Since this mechanism is mechanical, it would be present in the elastic range captured by the low pressure cubical triaxial test.

While there are methods to evaluate particle shape and surface roughness quantitatively, these methods result in highly uncertain values. Therefore, particle shape and surface roughness are evaluated qualitatively using microscopy. Particle shape and surface roughness are evaluated at 50x and 100x, respectively.



Figure 5-7 – Lodgepole pine particle shape (50x)



Figure 5-8 – Red oak particle shape (50x)

Comparing Figures 5-7 and 5-8, lodgepole pine particles are generally less angular and less fibrous than red oak fibers. Angularity is essentially the “sharpness” of the particle. Figure 5-8 displays the pointed ends of the red oak fibers, whereas lodgepole pine particles are rounder. Also, the lodgepole pine particles are more consistent in shape, nearly all particles, regardless of size, are rodlike with rounded ends. By contrast, oak fibers are more irregular; some are rectangular in shape while others are needlelike. Generally, the more irregular in shape and more angular a particle, the greater the mechanical interlocking effect. Surface roughness also plays a role in mechanical interlocking behavior.



Figure 5-9 – Lodgepole pine surface roughness (100x)



Figure 5-10 – Red oak surface roughness (100x)

In comparing Figures 5-9 and 5-10, red oak appears to have more surface contour than lodgepole pine, i.e. red oak appears to be rougher than the pine specimen. Red oak particles are jagged and protuberant, whereas lodgepole pine particles are smooth and dowel-like. Rougher surfaces tend to amplify bonding mechanisms such as Van der Waals forces and mechanical interlocking, since more surface area is in contact between particles. This observation, coupled with the more fibrous, angular, and irregular shape of red oak, likely serves to explain the dampened elastic response and greater bulk modulus observed in cubical triaxial testing. Generally, as particle size distributions become more irregular and rougher, bulk modulus increases while elastic response decreases.

By nature, irregular particle distributions are highly variable. This includes variability in shape and surface characteristics. Since microscopy is only capable of capturing small portions of the distribution, additional figures chosen to represent the range of observed shape and surface characteristics for both red oak and lodgepole pine are presented in Appendix E.

Chapter 6

Conclusions

Pellet producers are often limited by tree species due to geographic constraints. Often, producers have difficulty making pellets from different tree species. In reviewing the literature, the only difference in pelleting parameters that would vary with tree species is lignin content. By producing pellets from pure species across a range of lignin contents and subsequently producing pellets by mixing two species across this same range, the effect of lignin content on pelleting behavior between tree species is observed. Pellet durabilities ranged from very low, 30%, for the pure pine sample at 10% MC, to high, 95%, for the red oak sample at 10% MC. The major

conclusion of this work is that lignin content, within the range observable between various tree species, has no discernable impact on pellet durability. In the absence of this relationship, possibilities for other parameters that may vary between species and impact pelleting behavior were investigated.

Bulk materials are both difficult to characterize and relatively poorly understood due to their irregular shape and size distributions. However, the powder mechanics and pharmaceuticals fields have vast experience in characterizing and predicting the behavior of irregular particle distributions. The cubical triaxial testing method is borrowed from these disciplines in order to better explain the compaction behavior of sawdust. Specifically, bulk modulus and elastic response of the best and worst performers are investigated.

Lodgepole pine produced the least durable pellets, whereas red oak pellets were the most durable. The bulk modulus of red oak samples was much higher than that of lodgepole pine. This is thought to affect the pelleting process through the amount of friction experienced in the die. Harder materials will generate more friction, and thus more heat, during the compaction process. Heat is necessary to induce plastic deformation experienced by the feedstock as it passes the glass transition temperature and moves from the elastic to the plastic regime. This can also explain the fact that western producers, i.e. softwood producers, usually employ deeper dies than hardwood producers.

Further, the elastic response of the feedstock can be important in pelleting behavior. More elastic materials, those that tend to spring back more when pressure is released, will tend to expand upon exiting the die, resulting in cracks within the pellet. Lodgepole pine samples were observed to be much more elastic than red oak samples. This elastic response can be overcome

by solid bridges formed by the melting of natural binders within the material, but can be detrimental to pellet formation when these solid bridges are not as strong .

While lignin content varies within tree species, it was not observed to have an impact on pellet durability; however, significant differences in bulk material properties between pellet feedstocks that produced the highest and lowest durability values were observed. By looking at the bulk modulus and elastic response of densification feedstocks, among other bulk material properties, the biomass densification industry may gain insight into densification behavior. This is of particular importance as the industry looks to expand to feedstocks beyond woody biomass.

Chapter 7

Recommendations for Future Research

Biomass compaction behavior remains a relatively poorly understood process. Pellet producers talk of densification as more of an “art” than a science. While this is currently true, surely scientific phenomena can explain this process. This presents vast research opportunities in the biomass compaction arena.

One such opportunity highlighted in this research is the investigation of the related effects of bulk material response and particle characteristics on compaction behavior. The powder technology field also investigates the compaction behavior of irregular bulk materials, and has made significant headway in predicting powder compaction behavior by looking at the bulk material and individual particle properties that govern the compaction process.

Two bulk material properties highlighted here are the bulk modulus and elastic response of the bulk. These properties are only touched upon as indicators of compaction performance, and

further investigation into their predictive ability is warranted. Further, other bulk material properties, such as internal angles of friction and spring-back index, may be better predictors of compaction behavior, and should also be investigated.

Bulk material properties are inevitably the product of interparticle reactions. Therefore, when investigating bulk response, it is helpful to look to individual particle characteristics for explanation. Two properties, particle shape and surface roughness, were investigated only qualitatively by this work. Future efforts should attempt to characterize these properties, both qualitatively and quantitatively.

The powder compaction field has made large advancements in predicting the compaction behavior of irregular particle distributions by examining bulk material and particle characteristics. Biomass densification is very similar to powder compaction in that both applications look at the behavior of irregular particle distributions under high loads, and the biomass densification community would be well-served to look to the powder compaction field in identifying future research opportunities.

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Appendix A
PFI Standards

Table A-1 – PFI Fuel Grade Requirements

Fuel Property	Residential/Commercial Densified Fuel Standards - See Notes 1 - 9			
	PFI Super Premium	PFI Premium	PFI Standard	PFI Utility
Bulk Density, lb./cubic foot	40.0 - 46.0	40.0 - 46.0	38.0 - 46.0	38.0 - 46.0
Diameter, inches	0.250 - 0.285	0.250 - 0.285	0.250 - 0.285	0.250 - 0.285
Diameter, mm	6.35 - 7.25	6.35 - 7.25	6.35 - 7.25	6.35 - 7.25
Pellet Durability Index	≥ 97.5	≥ 97.5	≥ 95.0	≥ 95.0
Fines, % (at the mill gate)	≤ 0.50	≤ 0.50	≤ 0.50	≤ 0.50
Inorganic Ash, % - See Note 1	≤ 0.50	≤ 1.0	≤ 2.0	≤ 6.0
Length, % greater than 1.50 inches	≤ 1.0	≤ 1.0	≤ 1.0	≤ 1.0
Moisture, %	≤ 6.0	≤ 8.0	≤ 8.0	≤ 10.0
Chloride, ppm	≤ 300	≤ 300	≤ 300	≤ 300
Ash Fusion - See Note 8	NA	NA	NA	NA
Heating Value - See Note 1	As-Rec. ± 2SD	As-Rec. ± 2SD	As-Rec. ± 2SD	As-Rec. ± 2SD

Table 1 Notes:

1. There is no required value or range for Heating Value. It is required to print the mean higher heating value in BTU per pound as well as the ash content on the fuel bag label using a bar scale to represent the mean value ± 2 Std. Dev. See note 9.
2. The bag must be labeled indicating which PFI grade of material is in the bag. See note 9.
3. The bag label must also disclose the type of materials as well as all additives used. For purposes of this standard specification, additives are defined in 3.1.10. See note 9.

4. It is required that manufacturers include on their bags the PFI logo and in a printed block the guaranteed analysis of the fuel. See note 9.
5. PFI prohibits the use of any chemically treated materials. For purposes of this standard specification, chemically treated materials are defined in 3.1.11.
6. The following applies to all limits in this table: For purposes of determining the fuel grade, all properties must fall at or within the specified limits listed for a particular grade. Observed or calculated values obtained from analysis shall be rounded to the nearest unit in the last right-hand place of the figures used in expressing the limit in accordance with ASTM E 29-06b *Standard Practice for Using Significant Digits in Test Data to Determine Conformance with Specifications*.
7. It is the intent of these fuel grade requirements that failure to meet any fuel property requirement of a given grade does not automatically place a fuel in the next lower grade unless it meets all requirements of the lower grade.
8. It is required to report ash fusion properties at a frequency as specified in the PFI *Quality Assurance/Quality Control (QA/QC) Program for Residential/Commercial Densified Fuels*.
9. Refer to PFI *Quality Assurance/Quality Control (QA/QC) Program for Residential/Commercial Densified Fuels* for specific labeling requirements for fuel properties and other information.

Appendix B
Feedstock Particle Size Distributions

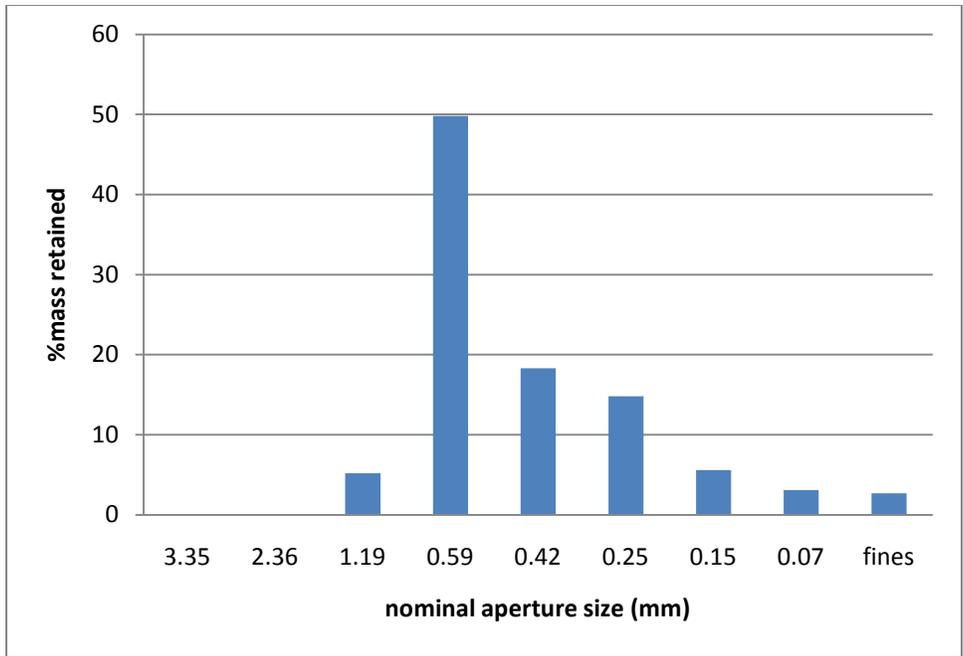


Figure B-1 – Cedar/Douglas Fir Particle Size Distribution

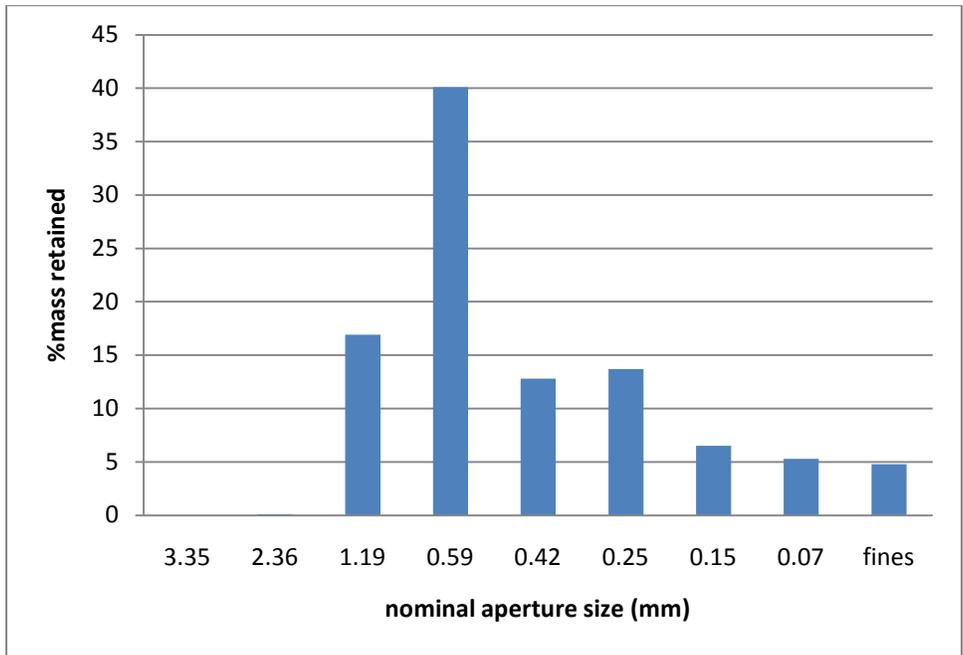


Figure B-2 – Lodgepole Pine Particle Size Distribution

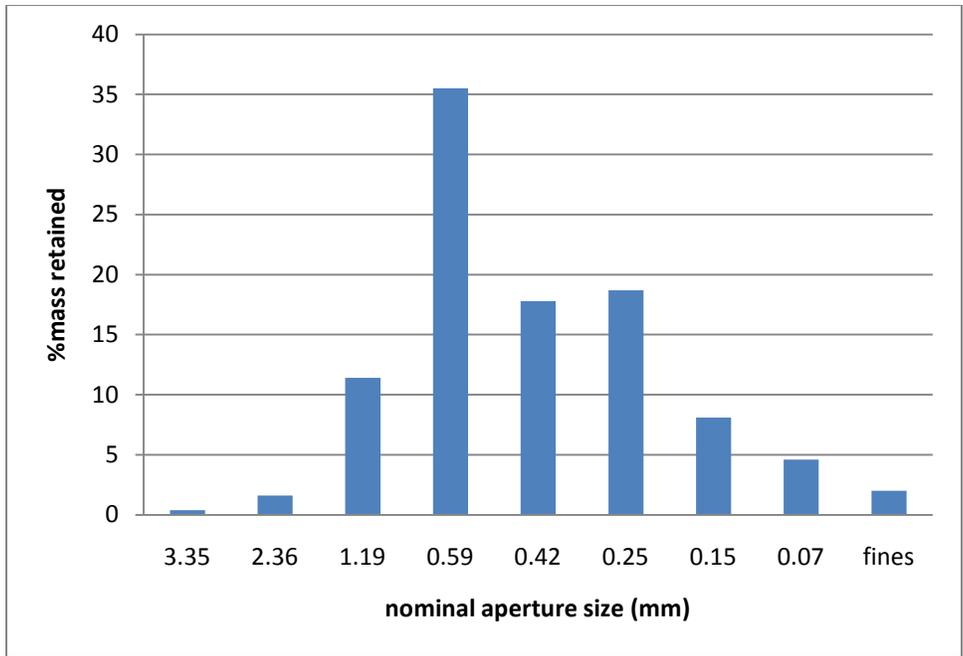


Figure B-3 – Red Oak Particle Size Distribution

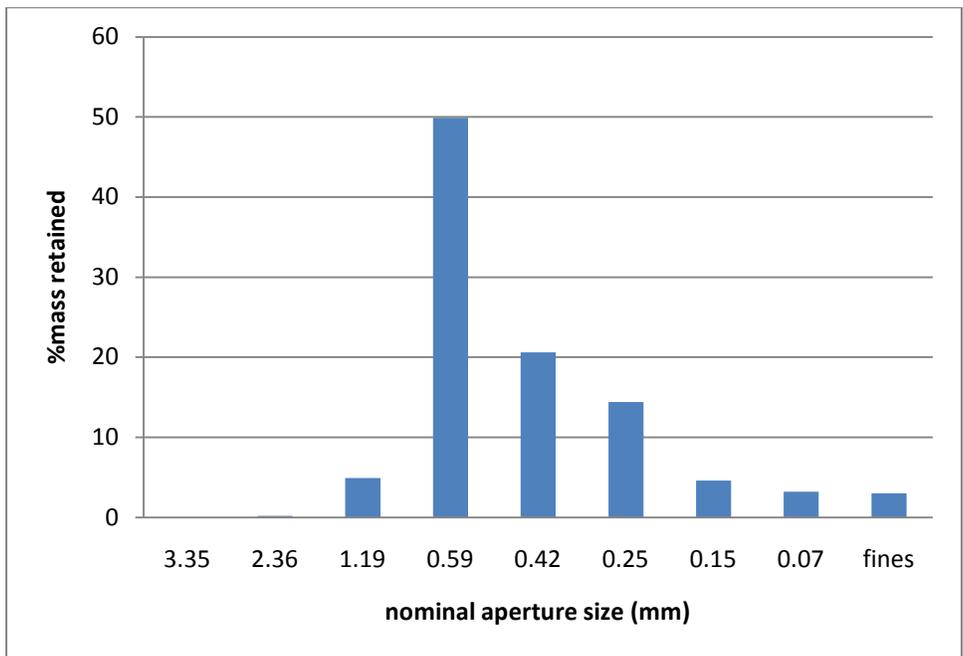


Figure B-4 – Walnut Particle Size Distribution

Appendix C
Cubical Triaxial Test Results

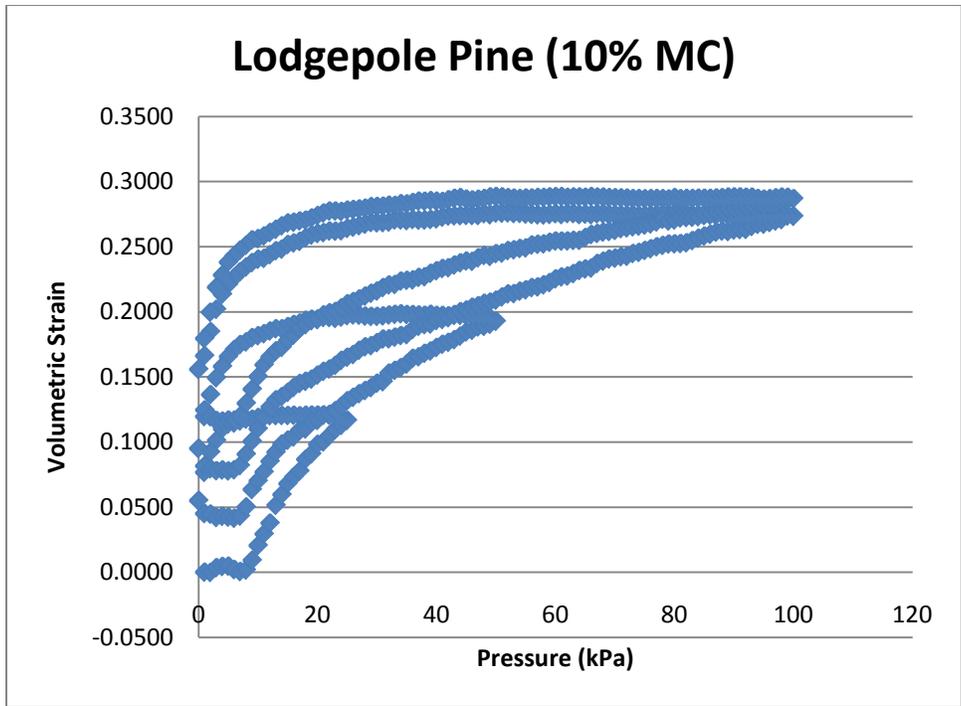


Figure C-1 – Volumetric Strain vs. Pressure for Lodgepole Pine (10% MC)

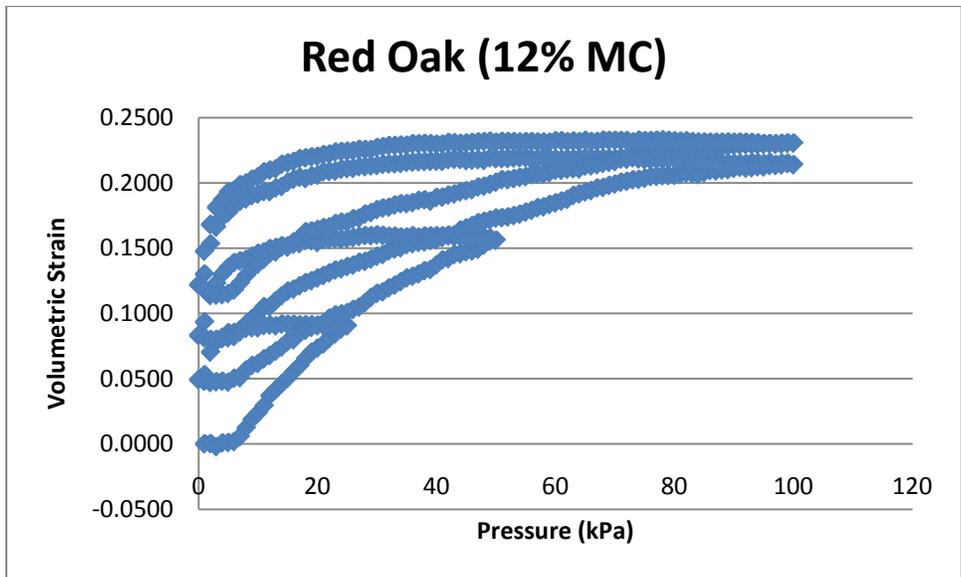


Figure C-2 – Volumetric Strain vs. Pressure for Red Oak (12% MC)

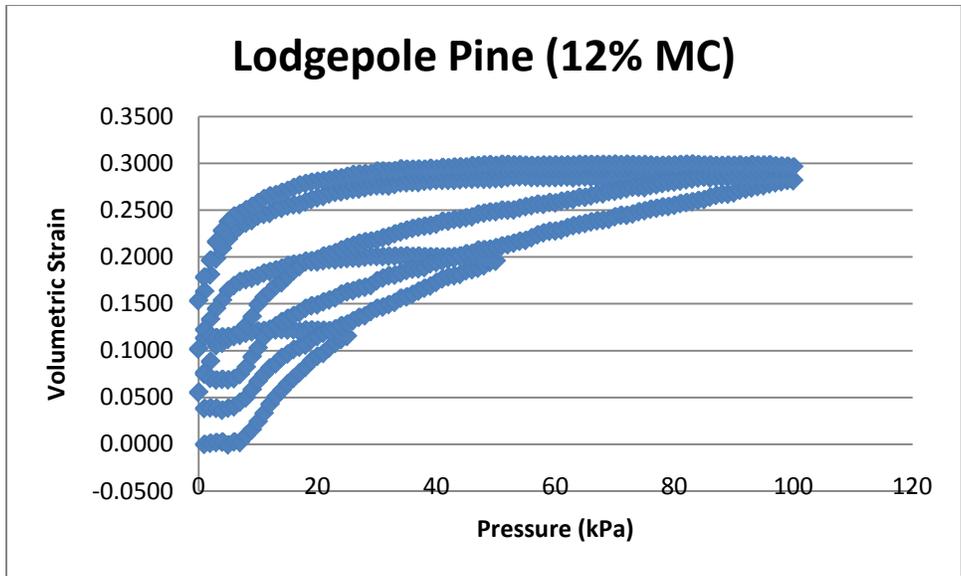


Figure C-3 – Volumetric Strain vs. Pressure for Lodgepole Pine (12% MC)

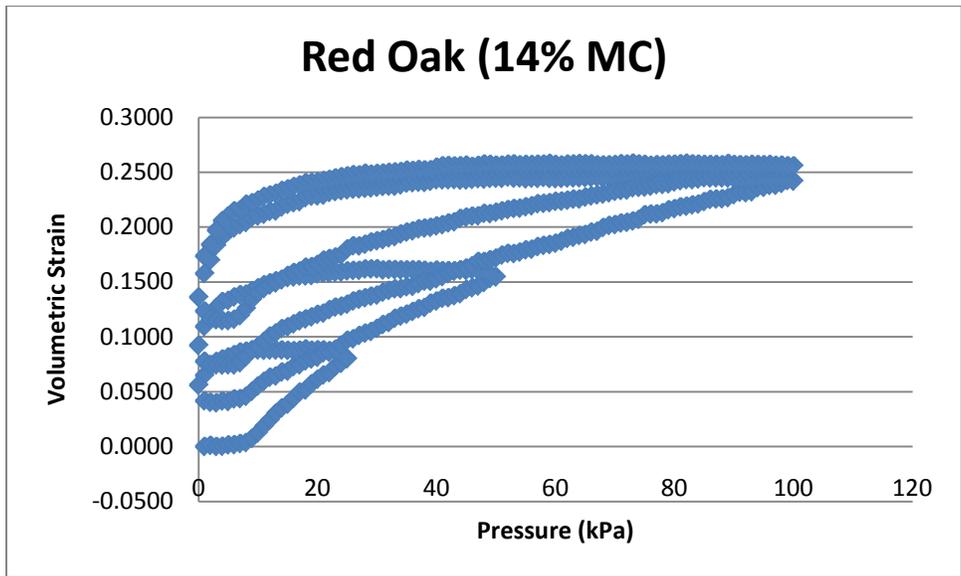


Figure C-3 – Volumetric Strain vs. Pressure for Red Oak (14% MC)

Appendix D

Lignin Content, Bulk Density, and Durability Values

Table D-1 – Sample Characterization

Sample	Moisture (% w.b.)	Lignin Content (%)	ρ_b (lb/ft ³)	PDI
specie(s)				
cedar/dougfir	10	30.17	45.8	87.24%
cedar/dougfir	12	30.17	46.3	94.52%
cedar/dougfir	14	30.17	44.8	93.64%
lodgepole pine	10	28.95	38.7	52.79%
lodgepole pine	12	28.95	43.0	89.26%
lodgepole pine	14	28.95	40.9	88.98%
maple	10	25.05	46.9	91.16%
maple	12	25.05	45.9	94.48%
maple	14	25.05	45.1	94.74%
red oak	10	25.77	49.6	95.04%
red oak	12	25.77	44.8	93.30%
red oak	14	25.77	44.6	93.66%
walnut	10	26.60	45.6	86.90%
walnut	12	26.60	46.6	88.96%
walnut	14	26.60	41.0	87.24%
Pine:oak mix ratio				
0:4.5	10	26.41	43.2	67.5%
0:4.5	12	26.41	45.0	90.3%
1.5:3	10	27.67	40.7	60.1%
1.5:3	12	27.67	43.3	93.2%
2.25:2.25	10	28.31	38.7	43.7%
2.25:2.25	12	28.31	42.1	86.3%
3:1.5	10	28.94	37.9	34.8%
3:1.5	12	28.94	-	-
4.5:0	10	30.21	38.5	29.6%
4.5:0	12	30.21	41.0	70.5%

Appendix E
Particle Shape and Surface Roughness Figures



Figure E-1 – Red oak surface roughness 1



Figure E-2 – Red oak surface roughness 2



Figure E-3 – Lodgepole pine surface roughness 1



Figure E-4 – Lodgepole pine surface roughness 2



Figure E-5 – Red oak particle shape 1



Figure E-6 – Red oak particle shape 2



Figure E-7 – Lodgepole pine particle shape 1



Figure E-8 – Lodgepole pine particle shape 2