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A Technical Review on Biomass Processing: Densification, Preprocessing, Modeling, and Optimization

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Jaya Shankar Tumuluru
Christopher T. Wright
Kevin L. Kenney
J. Richard Hess

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2950 Niles Road, St. Joseph, MI 49085-9659, USA
269.429.0300 fax 269.429.3852 hq@asabe.org www.asabe.org

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A Technical Review on Biomass Processing: Densification, Preprocessing, Modeling and Optimization

Jaya Shankar Tumuluru, Christopher T. Wright, Kevin L. Kenney and J. Richard Hess

Biofuels and Renewable Energy Technologies Department,
Energy Systems & Technologies Division,
Idaho National Laboratory, 2525 North Fremont Ave., Idaho Falls Idaho 83415
E mail ID: JayaShankar.Tumuluru@inl.gov

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Abstract. *Biomass from plants can serve as an alternative renewable and carbon-neutral raw material for the production of bioenergy. Low densities of 40–60 kg/m^3 for lignocellulosic and 200–400 kg/m^3 for woody biomass limits their application for energy purposes. Prior to use in energy applications these materials need to be densified. The densified biomass can have bulk densities over 10 times the raw material helping to significantly reduce technical limitations associated with storage, handling and transportation. Pelleting, briquetting, and other extrusion processes are commonly used methods for densification. The aim of the present research is to develop a comprehensive review of biomass processes including densification, preprocessing, modeling and optimization. Specific objectives include performing a technical review on (a) mechanisms of particle bonding during densification; (b) methods of densification including extrusion, briquetting, pelleting, and agglomeration; (c) effects of process and feedstock variables on biomass chemical composition and densification (d) effects of preprocessing (e.g., grinding, preheating, steam explosion, and torrefaction) on biomass quality and binding characteristics; (e) models for understanding compression characteristics; and (f) procedures for response surface modeling and optimization.*

Keywords. *Biomass, densification, pelleting, briquetting, extrusion process, densification process variables, modeling, optimization.*

Introduction

After coal and oil, biomass stands as the third-largest energy resource in the world (Bapat et al., 1997). Until the mid-19th century, biomass dominated global energy consumption. With steep increase in fossil fuel usage, biomass consumption for energy purposes has declined sharply for the past 50 years, but still provides about 1,250 million tons of oil equivalents (Mtoe) and supplies about 14% of the world's annual energy consumption (Purohit et al., 2006; Werther et al., 2000; and Zeng et al., 2007). Out of the 230 exajoules of estimated global primary energy, 56 exajoules—nearly one-fourth of the global primary energy—is used for agricultural practices (WEC 1994). Wood, agricultural stovers, and grasses are the most prominent biomass energy sources. Biomass, if properly managed, offers many advantages, the most important being that it is renewable and sustainable, and can significantly reduce net carbon emissions when compared with fossil fuels. Biomass is considered to be a clean development mechanism (CDM) for reducing greenhouse gas (GHG) emissions (Li and Hu, 2003).

The cheapest biomass sources are the waste products from wood or agro-processing units, but their supply is limited. To overcome this limitation, countries around the world are planting biomass crops for energy purposes. Most developed and industrialized nations are looking into developing technologies to use biomass more efficiently. In the USA and most of Europe biomass is already a competitive resource for energy production. USA and Sweden obtain about 4% and 13% of their energy, respectively, from biomass (Hall et al., 1992). In fact, Sweden has decided to phase out nuclear plants, reduce fossil fuel energy usage, and increase the use of biomass for energy (Björheden, 2006).

One of the major limitations of biomass for energy is its low density, typically ranging from 60–80 kg/m^3 for agricultural straws and grasses and 200–400 kg/m^3 for woody biomass like wood chips (Sokhansanj and Fenton, 2006; Mitchell et al., 2007). These low densities often make biomass material difficult to store, transport, and utilize. When this type of low density biomass is co-fired with coal, the difference in density between biomass and coal causes problems in feeding the fuel into the boiler and reduces burning efficiencies. To overcome this limitation, the density of biomass needs to be increased. Commercially, densification of biomass is performed using pellet mills and other extrusion processes, or briquetting presses, to increasing the density by about tenfold and help overcome feeding, storing, handling, and transporting problems.

The aim of this research is to develop a comprehensive review of biomass processing, which includes densification, preprocessing, modeling and optimization. Specific objectives include performing a technical review on (a) mechanisms of particle bonding during densification; (b) methods of densification including extrusion, briquetting, pelleting, and agglomeration; (c) effects of process and feedstock variables on biomass chemical composition and densification (d) effects of preprocessing (e.g., grinding, preheating, steam explosion, and torrefaction) on biomass quality and binding characteristics; (e) models for understanding compression characteristics; and (f) procedures for response surface modeling and optimization.

Densification

Conventional biomass densification processes can be classified into baling, pelletization, extrusion, and briquetting, which are carried out using a bailer, pelletizer, screw press, piston press, or roller press. Pelletization and briquetting are the most common densification used for solid fuel applications. These high pressure compaction technologies, also called binderless technologies, are usually carried out using either a piston press or a screw press (Sokhansanj et al., 2005). In a screw press, the biomass is extruded continuously through a heated tapered die. The quality of the extruded logs and the production process of a screw press are superior compared to piston press technology. However, compared to the wear of parts of a piston press, like the ram and die, screw press parts need more maintenance. The central hole in the densified logs produced by a screw press helps to achieve uniform and efficient combustion, and help to carbonize the material more quickly due to better heat transfer. Many researchers have studied the densification of woody and lignocellulosic biomass using a screw press, pellet mill and briquette press. Tabil and Sokhansanj (1996) worked on understanding the compression characteristics of alfalfa pellets; Ndiema et al. (2002) examined the influence of die pressure on relaxation characteristics of briquetted biomass; Adapa et al. (2002 & 2003) studied pelleting fractionated alfalfa products; Li and Liu (2000) investigated high-pressure densification of wood residues for fuel; Mani et al. (2006) studied the compaction characteristics of lignocellulosic biomass using an Instron; and Tumuluru et al. (2010) studied the effect of pelleting process variables on the quality attributes of wheat distiller's dried grains with solubles.

Mechanisms of Bonding of Particles during Densification of Biomass

The quality of the densified biomass depends on number of process variables like die diameter, die temperature, pressure, binder usage, and biomass preheating. Tabil and Sokhansanj (1996a & b) in their documents suggested that the compaction of the biomass grinds during pelletization can be due to the elastic and plastic deformation of the particles at higher pressures. According to their study, the two important aspects to be considered during pelletization are (1) the ability of the particles to form pellets with good mechanical strength and (2) the ability of the process to increase the density. The first aspect details the fundamental behavior of bonding or interlocking mechanisms that result in better densified biomass. Fig.1 shows the deformation mechanism of the powder particles during compression.

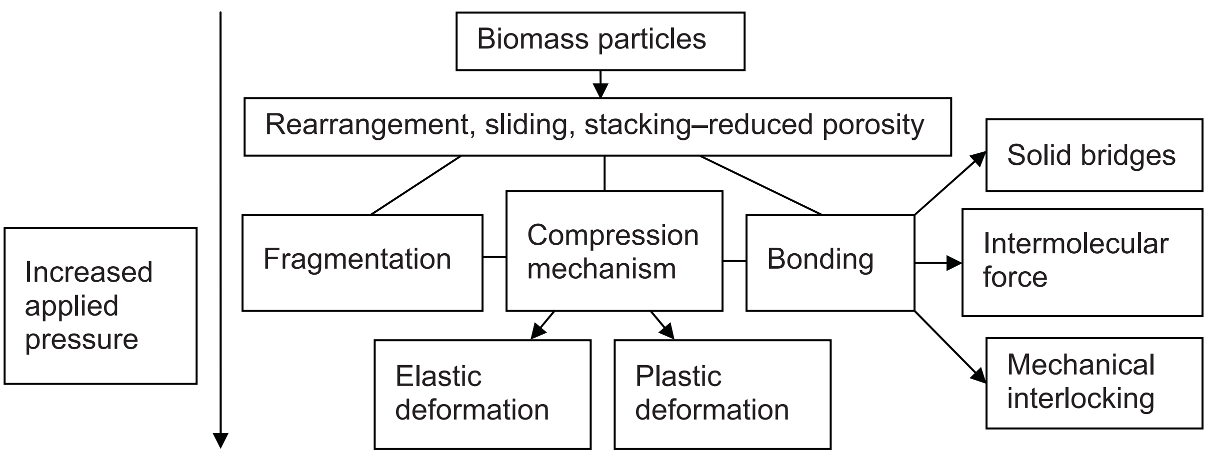


Figure. 1. Deformation mechanisms of powder particles under compression (Comoglu 2007; Denny 2002).

Through their study of the compaction behavior of some biomass grinds, Mani et al. (2002) postulated that there are three stages of biomass densification. In the first stage, particles rearrange themselves to form a closely packed mass where most of the particles retain their properties and the energy is dissipated due to inter-particle and particle-to-wall friction. In the second stage, the particles are forced against each other and undergo plastic and elastic deformation that significantly increase the inter-particle contact promoting bonding through van der Waal's and electrostatic forces. In the third phase, higher pressures significantly reduce the volume until the density of the pellet approaches the true density of the component ingredients. By the end of the third stage, the deformed and broken particles can no longer change their position due to a decreased number of cavities and achieve 70% inter-particle conformity.

Rumpf (1962) and Sastry and Fuerstenau (1973) suggested that the possible mechanism of binding during agglomeration can be due to the formation of solid bridges. The pressure applied during densification also reduces the melting point of the particles and causes them to move toward one another, thereby increasing the contact area and creating a new equilibrium melting point (York and Pilpel, 1972; Pietsch, 1984). Presence of liquid-like water during pelletization results in interfacial forces and capillary pressures that increase the bonding of the particles.

The chemical composition of the biomass, which includes compounds like cellulose, hemicelluloses, protein, starch, lignin, crude fiber, fat, and ash, also affect the densification process. During compression at high temperatures, the protein and starch plasticizes and acts as a binder, which assists in increasing the strength of the pelletized product (Briggs et al., 1999). Many researchers support that during densification of starch rich biomass using an extrusion process like pelleting the presence of heat and moisture gelatinizes the starch and results in better binding (Wood, 1987; Thomas et al., 1998). Lignin present in the biomass acts as a glue and improve the binding characteristics of the biomass. High temperatures and pressures, which are normally encountered during densification processes, result in softening of the lignin and improve the binding ability of the biomass. The low thermosetting properties and low melting point (~140°C) helps lignin to take an active part in the binding phenomena (van Dam et al., 2004).

Densification Technology

Screw Compaction or Extrusion

The aim of compaction using an extruder is to bring the smaller particles closer so that the forces acting between them become stronger and provide more strength to the densified material. During screw extrusion, the biomass material moves from the feed port through the barrel and compacts against a die with the help of a pressure building rotating screw. This process also causes friction from the shearing of biomass. The combined effects of internal and external friction heat and high rotational speeds (~600 rpm) of the screw cause an increase in temperature of the biomass. This heated biomass when forced through the extrusion die forms briquettes or pellets with the required shape. If the die is tapered the biomass gets more compacted. If the heat generated within the system is not sufficient for the material to reach a pseudo plastic state for smooth extrusion, heat can be provided to the extruders from outside the system using either band or tape heaters (Grover and Mishra, 1996). Fig. 2 shows a typical extruder with different zones for processing of biomass. Table 1 shows the specifications of typical biomass heat logs produced using an extruder. The Processing of biomass using screw compaction involves the following mechanisms (Grover and Mishra, 1996):

Table 1. General specification of extrudate produced by the Shimada SPMM 850 extrusion press. (Shimada systems, England, UK.

Raw Material prior to Extrusion (hard or soft wood)	
Moisture Content	8%
Average Particle Size	2–6 mm
Bulk Density	200 kg/m^3
After Extrusion	
Moisture Content	4%
Bulk Density	1400 kg/m^3
Calorific Value	4870 kcal (8400 btu/lb)
Ash Content	0.35–0.5%

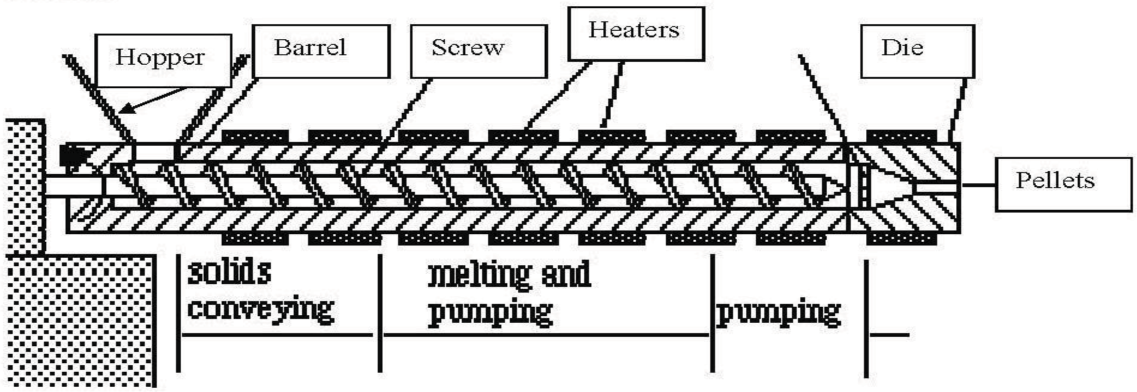


Figure 2. Typical representation of a screw extruder.
(Scientific Principles [http://matse1.mse.uiuc.edu/polymers/prin.html]).

1. Before reaching the compression zone which is solids conveying zone in Fig. 2 (a zone usually formed by tapering the barrel), the biomass gets partially compressed by the screw. This first stage of the process requires the maximum amount of energy in order for the particles to overcome friction.
2. Once the biomass is in the compression zone (melting and pumping zone in Fig. 2), the material becomes relatively soft due to high temperatures (200–250°C) and the material loses its elastic nature resulting in an increased area of inter-particle contact and local bridging. When the particles come closer, interlocking may occur. During its passage through the compression zone the biomass absorbs energy from friction causing heat to be uniformly mixed through the mass.
3. In the third stage, the biomass enters the tapering die (pumping zone in Fig. 2) where temperatures of approximately 280°C cause moisture to evaporated to steam. The steam in the tapering die zone helps condition the biomass as it is formed into a briquette.
4. In the final stage, the removal of steam and the final compaction take place simultaneously. This process results in a uniform pressure throughout the material helping form a uniformly dense briquette.

Briquette Machines

Densification of loose biomass using a briquette press is a viable and attractive solution to utilize biomass for fuel applications. Briquetting is usually performed using either hydraulic, mechanical, or roller presses. The densities of briquettes normally range from 900 to 1300 kg/m^3. The briquette is a clean, renewable fuel that can be used in furnaces, boilers, and open fires.

Unlike pellet mills, briquetting machines can handle larger sized particles with wider moisture contents without adding binders. In biomass briquetting processes, the material is compressed under high pressure and temperature causing it to self-bond a form a briquette due to thermoplastic flow. Lignin, which is a natural binder, is made available during the thermoplastic process resulting in the formation of high-density briquettes.

Hydraulic Piston Pump

Hydraulic piston presses are commonly used as briquetting machine for densification of biomass. The energy to the piston is transmitted from an electric motor via a high pressure hydraulic system. The throughput of a hydraulic press is lower than that of a mechanical press since the cycle of the cylinder is slower. In addition, the briquettes have a lower bulk density

(<1000 kg/m³) due to the fact that pressure is limited. However, these machines can tolerate higher moisture contents than the usually accepted 15% for mechanical piston presses (www.cfnielsen.com).

Mechanical Briquetting Press

Mechanical presses are typically used for large-scale production ranging from 200 kg/h to 2,500 kg/h. The mechanical press has a continuously rotating eccentric plunger that presses the raw material through a conical die. The required counter pressure of the press can only be adjusted by mounting a die with a different conicity. Since the mechanical press is electric driven and not hydraulic driven, energy losses are reduced and throughput efficiency is increased (www.cfnielsen.com). Table 2 compares specifications of a screw extruder and a piston press.

Table 2. Comparison of screw extruder and a piston press (FAO, 1996).

	Piston Press (Briquettes)	Screw Extruder (Extruded heat logs)
Optimum raw material moisture content	10–15%	8–9%
Wear of contact parts	Low for ram and die	High for screw
Output from machine	In strokes	Continuous
Power consumption	50 kWh/ton	60 kWh/ton
Density of briquette	1–1.2 g/cm^3	1–1.4 g/cm^3
Maintenance	High	Low
Combustion performance of briquettes	Not so good	Very good
Carbonization of charcoal	Not possible	Makes good charcoal
Suitability in gasifiers	Not suitable	Suitable
Homogeneity of briquettes	Not homogenous	Homogenous

Roller Press

Densification of biomass using a roller press works on the principle of pressure and agglomeration, where pressure is applied between two counter rotating rollers. The granular biomass, when forced through the gap between the two rollers, rotates with the rolls and is pressed in small dies or pockets (Yehia, 2007). Design parameters, which play a major role on the quality of the biomass, are the diameter of the rollers, minimum gap size, roll force, and shape of the die (Yehia, 2007).

Roller presses consist of two parallel cylinders of the same diameter that rotate on horizontal axes in opposite directions. The rotation of the rollers causes the feed to be drawn in from one side and ejected from the other in the densified form. The two rollers are separated by a small gap that can be adjusted based on biomass type and physical characteristics like particle size, moisture content, and binder addition. The final shape of the densified biomass depends on the type of die used (Yehia, 2007).

In the case of agglomerate production, by using smooth rolls the machine output can be a sheet having a specific thickness based on the gap provided between the rollers. The sheet produced is used to produce the agglomerates, as shown in Fig. 3. Whereas in the case of briquettes production dies with different configuration are used. The fines that are produced during briquetting using roller mills are recirculated back into the system as they help in improving the binding characteristics.

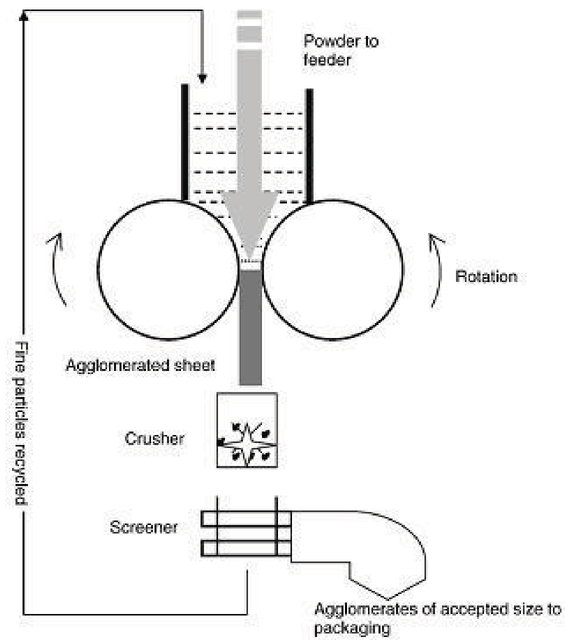
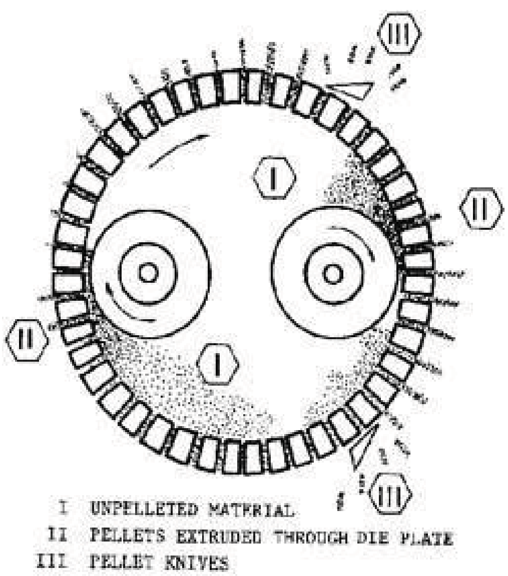


Figure 3. Roller press for agglomeration of biomass

Pellet Mill

Pelletizing is similar to briquetting except that it uses smaller dies (~ 30 mm) to produce smaller densified products called pellets. There are two main types of pellet presses: ring die and flat die. In general the die remains stationary and the rollers rotate. However, some pellet mills have dies that rotate and rollers that remain stationary during production. The die of a pelletizer is made of hardened steel that is perforated allowing the biomass to be forced through by the rotating die or rollers (Fig. 4). The various pellet mill components are shown in Fig. 5. Fig. 6 shows the dimensions of a commercial pellet mill die.



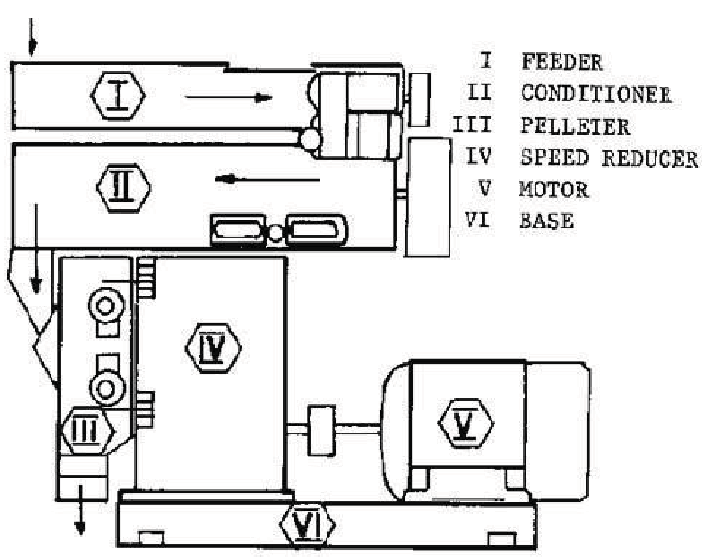
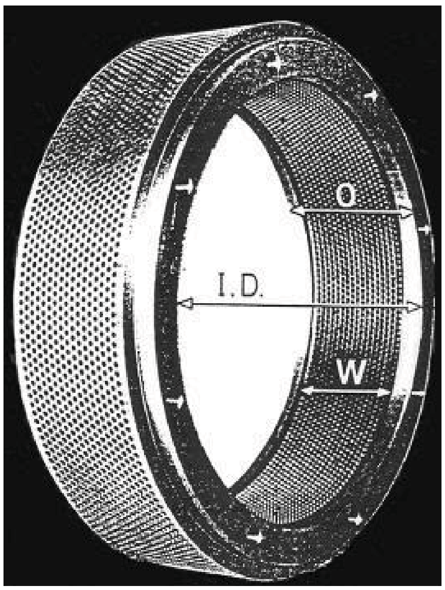


Figure 4. Working process of die.
Source: www.feedmachinery.com

Figure 5. Pellet mill components

In principle, the incoming feed from the feeder is delivered uniformly to the conditioner for the controlled addition of steam or binders such as molasses to improve the pelletization process. The various components of the screw conditioner are shown in Fig. 7. The feed from the conditioner is discharged over a permanent magnet and into a feed spout leading to the pelleting die. Steam is sometimes added to soften the feed and to partially gelatinize the starch, to form more durable pellets. As the die revolves, the friction-driven rollers force the feed through holes in the die. Cut-off knives mounted on the swing cover cut the pellets as they are extruded from the die. Unlike piston or screw presses, commercial pelletizers are not restricted by the density of the raw material having capacities in the range of 200 kg/h to 8000 kg/h and power consumption in the range of 15-40 kWh/ton (Grover and Mishra, 1996).



I.D. – Inside diameter of the die. This is the most common identifying factor for a die size.

O – Overall width of the die. There are normally two die widths for each die diameter.

W – Working width, measured between the two inside edges of the die grooves.

Grooves – Cut on the inside circumference of the die, into which the outside edges of the roll extend. This provides relief for the ends of the rolls so that the roll can be adjusted downward as the die wears away.

Die Working Area – Defined as the area between the two inside die grooves. This area is what is available for drilling the holes through which the pellets extrude.

(Source: Richard H. Leaver, Andritz Sprout, A division of Andritz Inc, Pennsylvania)

Figure 6. Dimensions of a commercial pellet mill die.

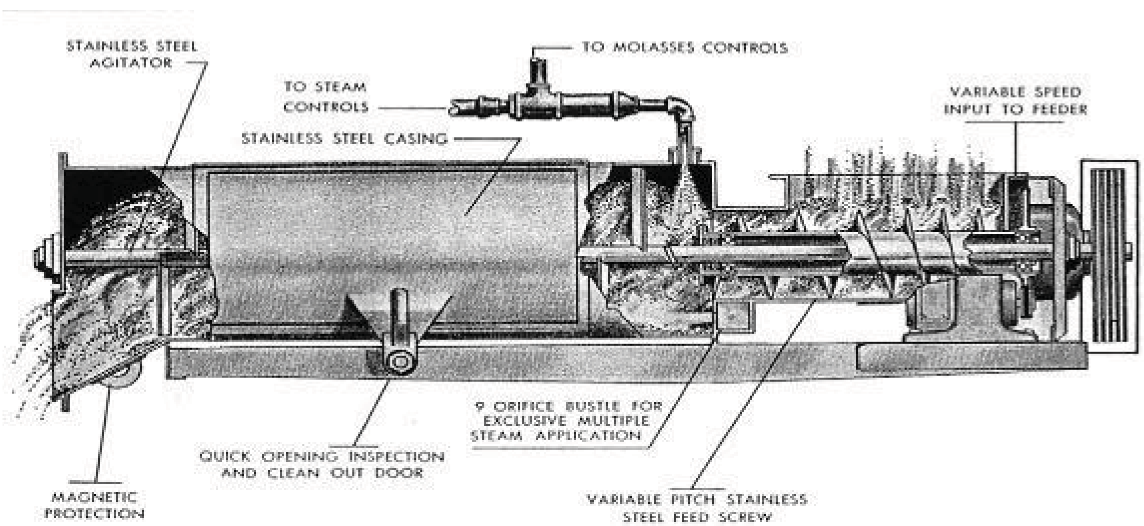


Figure 7. Component of a screw conditioner.
(Richard H. Leaver, Andritz Sprout, A division of Andritz Inc, Pennsylvania).

Agglomeration

Agglomeration is a method of size enlargement achieved by adhering biomass or chemical powder particles together. This technology can be applied to a variety of powders such as iron ores, fly ash, cement, hydrated lime, pulverized coal, and others. The application of agglomeration for biomass and charcoal is limited (Beaudequin, 1984; Reynieix, 1987). The most common agglomeration method is tumbling. Agglomeration equipment generally consists of a rotating barrel that is filled with metal balls of varying size. The rotation of the volume or agglomerator results in centrifugal, gravity, and frictional forces that cause a smooth rolling of the balls. These forces, together with inertial forces, press the balls against the powder, creating a pressure that forces them into balls (Siemons et al., 1989).

There are many design choices for an agglomerator. Choosing a balling device requires careful consideration of the particular application experienced in the field (Snow, 1984). The main process parameters for an agglomerator are ball residence time (depending on feed powder, feed rate, chamber volume, and pan-tilt angle) and proper rolling action (depending on scraper position, binder premixing, and pan-tilt angle).

Granulation-based agglomeration involves the following steps:

1. Fine raw material is continually added to the pan and wetted by a liquid binder spray
2. The disc's rotation causes the wetted fines to form small, seed-type particles (nucleation)
3. The seed particles "snowball" by coalescence into larger particles until they discharge from the pan.

Mort (2009) described the agglomeration process as a function of various material properties and process parameters, which are illustrated in Fig. 8. Figs. 9 and 10 show a disk pelletizer and rotary drum granulator which are commonly used in the agglomeration of powders.

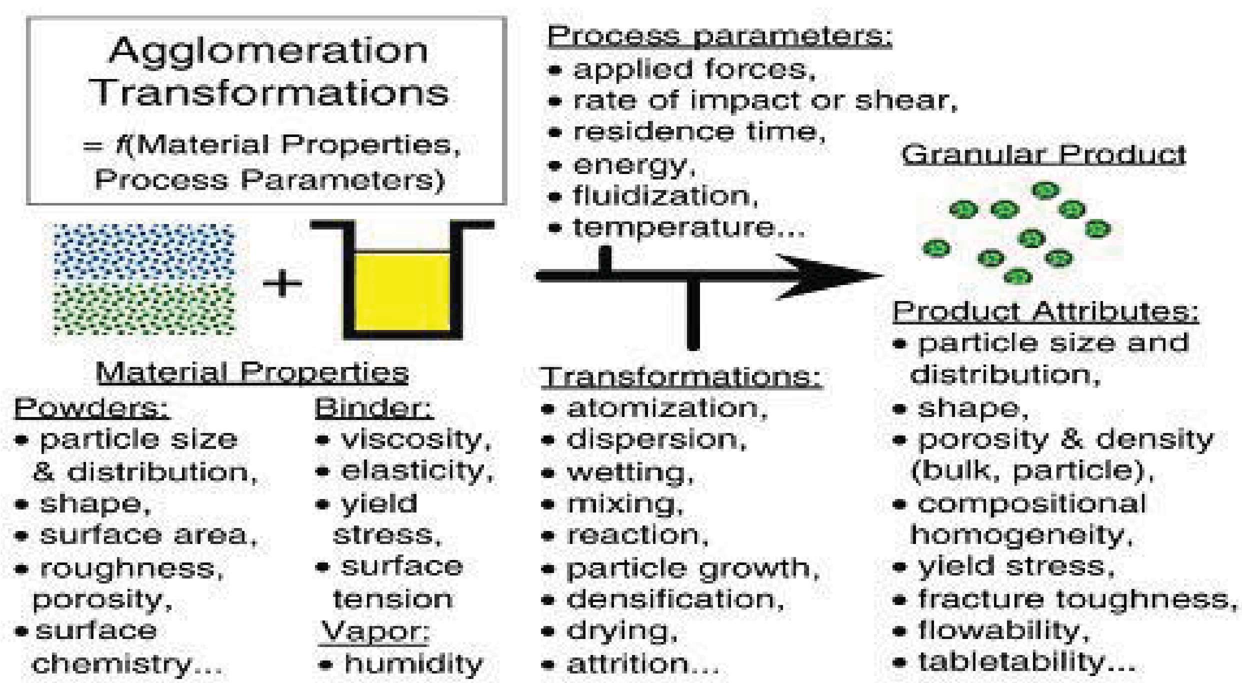


Figure 8. Agglomeration as a function of material properties and process parameters (Source: Mort (2009).

Figure 9. Disk pelletizer.

Figure 10. Rotary drum granulator.

Source: Feeco International (www.feeco.com)

Densification System Variables

Controlling the densification system variables is key for achieving the desired densified biomass quality. Specifically, the quality of the densified product can be managed by controlling conditions such the manufacturing process, changes in formulation, and the use of additives (MacMahon, 1984). In addition, process variables such as die thickness, retention time, roller-die ring gap (Wetzel, 1985), steam conditioning, and feed rate (MacMahon, 1984) affect the quality of densified biomass like density and durability. In studies on densification of biomass, Shaw (2008) identified that process variables (die temperature, pressure, and geometry) feedstock variables (moisture content and particle size and shape) and feedstock composition (protein, fat, cellulose, hemicelluloses and lignin) all impact the quality of the densified biomass.

Process Variables

Process variables (such as temperature, pressure, retention time, and die geometry and speed) play an important role in achieving the desired product quality of densified biomass in terms of durability, density, and calorific value.

Temperature

Quality attributes like durability and bulk density of densified material are significantly influenced by temperature. Hall and Hall (1968), in their studies on alfalfa and Bermuda grass densification, found that for a given moisture content, the pressure required to obtain a certain wafer density was reduced by the addition of heat in the die. In addition, the upper limits of moisture content at which a certain pressure was able to produce a specific wafer density was increased by the addition of heat. Hill and Pulkinen (1988) found that high-temperature conditioning of raw materials increased pellet durability. They found that the pellet durability of alfalfa increased by about 30–35% when the pelleting temperature was increased from 60 to 104ºC. Mani et al. (2003) and Sokhansanj et al. (2005) also observed that higher temperatures resulted in reduced resistance of the material against equipment components resulting in better quality pellets. Smith et al. (1977), in their study of briquetting wheat straw, found that the degree of compaction and dimensional stability went up as the temperature increased from 60 to 140°C. They also found that briquette expansion decreased when the die temperature was between 90 and 140°C. They further observed that briquettes were surface charred and slightly discolored at temperatures above 110°C due to chemical degradation. Tabil and Sokhansanj (1996) found that pelleting temperatures >90°C significantly improved durability values of alfalfa pellets. They concluded that it is necessary to precondition the grinds to above 90°C to achieve high pelleting temperatures that promote better bonding of particles and produce good durable pellets. In their study on corn stover and switchgrass, Kaliyan and Morey (2006) used the glass transition temperature of the biomass to understand the densification behavior. Their studies included three different temperatures: two within the glass transition temperature (75 and 100°C) and one outside (150°C). The durability values of the densified biomass outside the glass transition temperature were lower compared to ones within the range. The Glass transition temperature was found to be inversely related to moisture content.

Pressure

Pressure plays an important role on the quality of the pellets made from agricultural biomass. Butler and McColly (1959) observed that the density of chopped alfalfa hay pellets was proportional to the natural logarithm of the applied pressure and found that increasing pressure significantly increased the density. Yaman et al. (2000) recommended that briquetting pressure should be selected such that it influences the mechanical strength by increasing the plastic deformation. However, above an optimum briquetting pressure, fractures may occur in the briquette due to sudden dilation. For a given die size and storage condition, there is a maximum die pressure beyond which no significant gain in cohesion (bonding) of the briquette can be achieved (Ndiema et al., 2002). High pressures and temperatures during densification may develop solid bridges by a diffusion of molecules from one particle to another at the points of contact, which increases density. Li and Liu (2000) observed that compression of oak sawdust at pressure application rates varying from 0.24 to 5.0 MPa/s had a significant effect on the dry density of the product. In their article on the compaction of biomass waste materials like waste paper, Demirbas et al. (2004) observed that increasing the pressure from 300 MPa to 800 MPa on biomass with ~7% moisture content initially increased the density sharply, from 0.182 g/mL to 0.325 g/mL, and then the densities increased slightly to 0.405 g/mL.

Retention or Hold Time and Relaxation Time

The quality of briquettes is significantly influenced by the retention or hold times of the materials in the die (Tabil and Sokhansanj, 1996). However, Al-Widyan et al. (2002) found that the retention times between 5 and 20 seconds did not have a significant effect on olive cake briquette durability and stability. In their study on high pressure densification of wood residues to form an upgraded fuel, Li and Liu (2000) found that the hold time for oak sawdust had more effect at lower pressures than at higher pressures. At the highest pressure (138 MPa), the effect of holding time was negligible. Also, they observed that the holding time had little effect on the expansion rate. A 10-second holding time could result in a 5% increase in log density, whereas at holing times longer than 20 seconds, the effect diminished significantly.

In general, relaxation time has a significant effect on the density of the materials. Final relaxed density of briquetted fuel and the relaxation behavior following removal from the die depend on many factors related to die geometry, the magnitude and mode of compression, the type and properties of the feed material, and storage conditions. Many studies on high-pressure compaction of biomass materials have indicated that upon removal of densified material from the die, the density of the compacted material decreases with time to a final relaxed density. For most feed materials, the rate of expansion is highest just after the removal of pressure and decreases with time until the particle attains constant volume (Carre et al., 1987; Miles, 1980). The relaxation characteristics, which are mainly measured by the percentage of elongation and increase in voidance, depend on many factors related to the feed material and storage conditions, such as relative humidity (Wamukonya and Jenkins, 1995). Shrivastava et al. (1990) used statistical analysis for the results obtained for rice husks to establish a multiple correlation equation of the following form:

$$Y = \alpha_0 + \alpha_1 P + \alpha_2 T \tag{1}$$

Where

Y = percent volume expansion;

T (°C) and P (kg/m^2) = the die temperature and pressure, respectively and

α_0, α_1 and α_2 = constants.

Die Geometry and Speed

Die geometry refers to the size and shape of the die. These dimensions significantly affect the amount of material that can be pelleted and, the energy required for compression. Die geometry also influences product properties like moisture content, bulk density, and durability. The L/D ratio (length to diameter of the pellet) can be a good metric for the degree of compression during pelletization. An increase in pelletizing pressure increases the length of the pellet, whereas an increase in pellet diameter decreases the pelleting pressure. Hence, the dimensions of the die and the press channels in the matrix have a strong influence on determining the pressure needed to press pellets through the matrix. Butler and McColly (1959) found that for a constant mass of material, pellet density and length was greater for smaller diameter chambers at a given pressure, which resulted in less expansion. Tabil and Sokhansanj (1996) studied the effect of process parameters like steam conditioning, die geometry, L/D ratio, die speed, and particle sizes of the biomass and found that at higher conditioning temperatures (>95°C) the durability of the pellets increased. They also concluded that the durability of the pellets improved when a smaller die with higher L/D ratios was used. Shankar et al. (2005) found that die barrel temperature and screw speed significantly affected quality attributes like bulk density and hardness of the biomass feed mix during extrusion processing. Shankar et al. (2008) found that L/D ratio and screw speed significantly affected the flow rate of the biomass feed mix, and this flow behavior affects the final quality of the pelleted biomass. Hill and Pulkinen (1988) reported that the durability of alfalfa pellets increased by about 30–35% at an L/D ratio between 8 and 10. Heffner and Pfost (1973) evaluated the effect of three die sizes (4.8 × 44.5, 6.4 × 57.2 and 9.5 × 76.2 mm) on the durability of animal feeds. They found that pellets produced on the smallest die had the best durability values. In their study of distiller dried grains with solubles (DDGS), Tumuluru et al. (2010a), found that a larger die diameters of 7.2 mm produced less-durable DDGS pellets compared to a smaller die diameter of 6.4 mm, for both with and without the addition of steam.

Feedstock Variables

Feedstock variables include moisture content and particle size, shape, and distribution. As with process variables, these have a significant influence on pellet quality.

Moisture Content

Moisture in the biomass facilitates starch gelatinization, protein denaturization, and fiber solubilization processes during densification. Steam-heat-treated biomass is superior to raw biomass since the additional heat modifies these physiochemical properties to the extent that binding between particles is enhanced, resulting in improved physical pellet quality (Thomas et al., 1997). Mani et al. (2003) observed that moisture in the biomass during the densification process increases the bonding via van der Waal's forces, thereby increasing the contact area of the particle. Mani et al. (2006) found that low moisture content in the biomass (5–10%) resulted in denser, more stable, and more durable briquettes compared to those produced from biomass with higher moisture content (15%). Li and Liu (2000) recommended an optimum moisture content of ~8% to produce high-density briquettes. They also recommended a moisture content of 5–12% to produce good quality logs in terms of density and long-time storage properties from hardwood, softwood, and bark. They also noted that pellets or briquettes tend to become fragile in a few days if the moisture content is less than 4% due to moisture absorption from the environment. Sokhansanj et al. (2005) identified that feed material, which contains higher proportions of starch and protein, will produce more durable and higher quality pellets than biomass containing only cellulosic material. They also concluded that the optimum moisture content for pelleting cellulosic materials is 8–12%, whereas for starch and protein materials (mostly animal feeds), the optimum moisture content can range up to 20%.

Particle Size, Shape, and Distribution

In general, the density and durability of pellets is inversely proportional to the particle size because the smaller particles have greater surface area contact during densification. MacBain (1966) and Payne (1978) concluded that medium or fine-ground feed constituents are desirable in pelleting because these sizes have greater surface area for moisture addition during steam conditioning, which increases starch gelatinization and promotes better binding. They also reported that a certain percentage of fines to medium particles are required in commercial pellet mills to improve pelleting efficiency and reduce pelleting costs. However, very small particle sizes can lead to jamming of the pellet mills and affect production capacity.

Table 3 indicates the particle size distribution for producing good quality pellets (Payne, 1996). Smith et al. (1977) mentioned that the compaction and stabilization of straw may be different than grasses due to the fact that straw tends to have significantly smaller leaf content. Hill and Pulkinen (1988) reported for alfalfa pellets that an increasing screen size from 2.8–6.4 mm reduced the durability of the alfalfa pellet by more than 15%. They also reported that particle size and pellet durability must be balanced against the energy required to accomplish size reduction.

Table 3. Optimum particle size distribution for producing quality pellets from agricultural biomass.

Sieve size (mm)	Material retained on sieve
3.0	≤1%
2.0	≤5%
1.0	≈20%
0.5	≈30%
0.25	≈24%
<0.25	≥20%

Biomass Composition

Feedstock composition is one of the major variables that contributes to the quality of densified materials. Raw biomass has both low molecular weight and macromolecular compositions. Low molecular weight substances include organic matter, inorganic matter and macromolecular substances include cellulose, hemi-cellulose, and lignin (Mohan et al., 2006). Understanding some of the major compositional changes that takes place due to reactions during biomass processing can be useful in understanding their compaction behavior. Thomas et al. (1998), identified some of the important ingredients that influence the pellet quality, including starch, protein, sugar and non-starch polysaccharides (NSP), fat, fiber, inorganic matter, and water.

Starch

Starch is a D-glucose polymer with branched (amylopectin) or un-branched (amylose) chains (Collado and Corke, 2003). The behavior of starch is mainly controlled by the gelatinization process it undergoes at high processing temperatures. Starch granules at higher temperature and moisture contents influence the textural properties of many foods and feeds (Shankar and Bandyopadhyay, 2006). Collado and Corke, 2003 stated that the starch undergoes gelatinization, pasting, and retrogradation reactions, of which gelatinization plays the major role during pelletization. Gelatinization of starch is an irreversible process and is mainly influenced by the densification process variables like heat, water, shear, and residence time (Thomas et al., 1999). The non-food applications of starches include adhesives for board, paper and labels in the paper industry (Thomas et al., 1999). In the pharmaceutical industry starch is widely used as a binder or filler in tablet formulations (Alebiowu and Itiola, 2002). During the feed and food pelleting process, starch not only acts as a binder but also as a lubricating agent, and helps to easy the flow of materials through a pellet mill or an extruder.

Protein

Protein in the biomass is heated during the densification process and undergoes denaturization leading to the formation of new bonds and structures with other protein, lipids and starch available in biomass and improves the binding capacity (Thomas et al., 1998 and Nyanzi and Maga, 1992). According to Briggs et al. (1999) and Wood (1987), increasing the protein content increases the pellet durability. In addition, using raw protein during pelletization of biomass improves the physical quality of the pellets compared to denatured proteins. Tabil (1996) reported that if there is sufficient natural protein in the biomass during pelletization it will plasticize under heat and improve the binding properties.

The interactions between starch and proteins also have significant influence on the densification process and quality of the densified biomass. Sokhansanj et al. (2005) reported that feed material with larger fractions of starch and protein composition produced denser and more stable pellets than biomass with larger composition of cellulose. Shankar et al. (2008) and Shankar and Bandyopadhyay (2004 & 2005) found that fish content of the feed, barrel temperature, and feed moisture played an important role in the hardness of the extrudates. They observed that at higher processing temperatures (100–200°C) the cross-linking of protein, starch and lipids resulted in more durable and stable pellets. Other studies by these authors using a scanning electron microscopy support impact of protein cross-linking with the other constituents like starch and lipids. In his article on functional properties of the protein and starch fractions, Wood (1987) concluded that protein has a significant influence on feed quality attributes like hardness and durability.

Lipid/Fat

The fat added to the feed biomass acts as a lubricator during pelletization and increases the throughput of the pellet mill. However, an increase in fat decreases the binding capacity and may require additional binders to improve its durability and hardness. Shankar et al. (2008), in their article on extrusion of aquafeed with high lipids of about 8–10%, found that a commercial binder is required to improve its density and water stability values. Briggs et al. (1999) found that increased oil content produced lower quality pellets because fat is hydrophobic in nature and may interfere with the binding of the feed particles during pelletization. In addition, if the fat is added during pelletization it acts as a lubricant between the particles and reduces the pelleting pressure (Thomas et al., 1998).

Lignocellulose

Lignocellulose is the descriptor of nonfood-related biomass such as trees, grasses, and biomass waste materials. Wood-based polymers are comprised of cellulose (40–60%), hemicelluloses (20–40%), and lignin (10–25%), and together are known as lignocellulosic material (United States, Department of Energy, 2006). Typical lignocellulosic content of some example plant materials is shown in Table 4.

Table 4. Typical lignocellulosic content of some plant materials (Mohan et al., 2006).

Plant material	Lignocellulosic content (%)		
	Hemicelluloses	Cellulose	Lignin
Orchard grass (medium maturity)[a]	40.0	32.0	4.7
Rice straw[b]	27.2	34.0	14.2
Birch wood[b]	25.7	40.0	15.7

a. Data taken from Van Soest (1964)
b. Data taken from Solo (1965)

Cellulose

Cellulose forms crystalline microfibrils that are surrounded by amorphous cellulose inside plant cells (Chen et al., 2004). The hemicelluloses and lignin form an amorphous matrix that reinforces the cellulose microfibrils. The structural integrity of the cellulose is produced by hydrogen bonding that occurs between the glucose monomers (Goldstein, 1981). According to Nelson and Cox (2005), cellulose (the tough, water insoluble substance found in cell walls, particularly in the stalks, stems or trunks, and woody portions of the plant body) is considered to be an abundant source of carbon in biomass. In their article on hot pressing of wood material, Zandersons et al. (2004) concluded that the binding strength of wood based products mainly depends on converting the cellulose to an amorphous state.

Hemicelluloses

The hemicelluloses found in the cell wall are a heteropolysaccharide, or a combination of many other sugars other than just glucose. The amorphous structure of hemicelluloses is due to branching and is more easily hydrolyzed or can be dissolved in alkali solution. Some researchers believe that natural bonding may occur due to the adhesive degradation products of hemicelluloses.

Lignin

Lignin is a random network polymer with a variety of linkages based on phenyl propane units (Zandersons et al., 2004). The lignin molecule in a plant provides many structural purposes such as acting like glue to the cellulose fibers. The presence of lignin in plant materials allows pelletization without adding binders. van Dam et al. (2004) reported that lignin exhibits thermosetting properties at working temperatures of >140°C and acts as an intrinsic resin in binderless board production. Lignin is the component that permits adhesion in the wood structure and acts as a rigidifying and bulking agent (Anglès et al., 2001). Lehtikangas (1999) stated that moistures of about 8–15% in biomass will reduce the softening temperature of lignin to 100–135°C by plasticizing the molecule chains. The adhesive properties of thermally softened lignin contribute considerably to the strength characteristics of briquettes made of lignocellulosic materials (Granada et al., 2002).

Binders used in Biomass Densification

Binders are normally used to reduce the wear on production equipment and to increase the abrasion resistance of the fuel. Binders also improve the binding characteristic of the biomass and produce more durable pellets. In pellet production, binders are allowed but need to be specified as part of the final product. The most commonly used binder in pellet making is lignosulphonates (Wafolin) (Tabil and Sokhansanj, 1997).

Lignosulfonates are commonly used as binders for animal feeds and have been considered the most effective (Anonymous, 1983; MacMahon, 1984). The composition includes sulfonate salts made from lignin from sulfite pulp mill liquors. The general quantity to include for effective binding ranges from 1–3%.

Bentonite is a common substance used as a binder in feed pelleting. Also referred to as colloidal clay, bentonite is an aluminum silicate composed of montmorillonite. During processing, the binder forms a gel with water and helps in improving the binding characteristics. Pfost and Young (1973) reported that the addition of bentonite at an inclusion rate of 100 kg/ton of feed mash significantly improved the durability of the poultry feed consisting of ground yellow corn, ground sorghum grain, and soymeal ingredients.

Starches are also used in the food industry as a thickener or a binder. Wood (1987) reported that precooked starch works as a good binder during pelletization. Proteins are considered natural binders. The heat developed as the material passes through the die can help produce more durable pellets due to interactions of proteins with other biomass compositions such as lipids and starches. Some agricultural biomass, such as alfalfa, has a high protein content and can be used as binder to improve the durability of the pellets made from lignocellulosic biomass.

Preprocessing of Biomass

Grinding

Prior to densification, the biomass is ground to a certain particle size. This grinding partially breaks down the lignin, increases the specific area of the material, and improves binding. Peleg (1977) and Peleg and Mannheim (1973) suggested that particle size has a significant effect on the binding characteristics. Fine powders have more contact points, exposed surface area, and surface energy per unit weight regardless of their physical and chemical characteristics. In his study of wheat and barley straws and corn stover, Mani et al. (2006) concluded that the particle size has significant effect on the mechanical properties of pellets.

Preheating

Preheating the biomass before densification is common because it results in a better quality product. Most commercial pellet or briquette producers use preheating to form stable and dense pellets or briquettes (Bhattacharya et al., 1989; Bhattacharya, 1993). Aqa and Bhattacharya (1992) indicated that preheating biomass could significantly increase the throughput of the pelletizing machine and reduce the energy requirement per kilogram of pellets formed.

Steam Conditioning and Explosion

Steam explosion is a technique that has been widely used and is an efficient method of pretreating lignocellulosic biomass prior to densification or ethanol production. In the steam explosion process, biomass is introduced into a reactor and heated under steam pressure for a short time producing significant physical, chemical, and structural changes and making more lignin sites available for binding during pelletization (Liu and Wyman 2005). During steam explosion, hemicelluloses become more water soluble, cellulose is slightly depolymerized, and lignin melts and gets depolymerized (Toussaint et al., 1991). Steam explosion breaks down the lignin into low-molecular weight products that retain the basic lignin structure and are moderately reactive. Mosier et al. (2005) postulated that the compression and compaction characteristics of the biomass can be improved by disrupting lignocellulosic biomass materials via steam explosion pretreatment.

In general, steam explosion makes biomass more susceptible to enzymatic hydrolysis. First, lignin is extensively depolymerized by cleavage of the β-aryl-ether bonds and is soluble in alkaline solutions or certain organic solvents. Second, hemicelluloses, which are predominantly soluble in water, are partially broken down and condense with lignin causing an increase in the lignin content. The major effect of a steam explosion is the large increase in the accessibility of the cellulose to enzymatic hydrolysis (DeLong, 1981; Foody, 1980). According to Zandersons et al. (2004) the activation of lignin and changes in the cellulosic structure during steam explosion help in the formation of new bonds, which in turn produces more durable pellets. In their article on pelleting of DDGS grinds using a pilot-scale pellet mill, Tumuluru et al. (2010a) reported that steam conditioning before pelletization produced higher durable pellets using a 6.4 mm die compared to a 7.4 mm die.

Torrefaction

Torrefaction is a method to improve the properties of the biomass for energy conversion. Felfli et al., 1998 define torrefaction as a slow heating of biomass in an inert or reduced environment to a maximum temperature of 300°C. Similarly, Zanzi et al., 2002 define Torrefaction as a group of products resulting from the partially controlled and isothermal pyrolysis of biomass occurring in a temperature range of 200–230ºC and 270–280ºC. Thus, the process can also be called a mild pyrolysis as it occurs at the lower end in terms of temperature of the pyrolysis process. The treatment yields a solid uniform product with lower moisture content and higher energy content compared to raw biomass. Most of the smoke-producing compounds and other volatiles are removed during torrefaction producing a final product that has approximately 70% of the initial weight and 80–90% of the original energy content (Arcate, 2000 & 2002). The torrefaction process opens up a number of lignin active sites by breaking down the hemicelluloses matrix and forming fatty unsaturated structures that help in binding. The bulk densities of torrefied pellets range from 750 to 850 kg/m^3 (Bergman and Kiel, 2005).

Torrefaction reduces the variability in feedstocks resulting from different types of biomass species, climatic and seasonal variations, storage conditions, and time (Lehtikangas, 1999). Torrefaction helps in developing a uniform-format material to produce high-quality densified biomass. Torrefaction affects the biomass physical properties like grindability, hydrophobicity, pelletization, and calorific value. During the torrefaction process, biomass loses its tenacious nature mainly through the breakdown of the hemicelluloses matrix and depolymerization of the cellulose, which results in a decrease in fibers length (Bergman et al., 2005; Bergman and Kiel, 2005 and Hakkou et al., 2006).

Torrefaction results in the shrinking of biomass and produces a light-weight, flaky, and fragile material that is easier to grind and pulverize (Arias et al., 2008). Studies conducted by Bergman and Kiel (2005) on grinding energy requirements of the raw and torrefied biomass reported that that torrefied biomass required significantly less power consumption (~70– 90%) and increased grinding capacity by about 7.5 to 15 ton/hr.

Torrefaction also improves aerobic stability of the processed biomass. Torrefied biomass is hydrophobic in nature and does not absorb moisture during storage or transportation. This occurs because the hydroxyl group (OH) is destroyed and the material cannot support the formation of hydrogen bonds (Pastorova et al., 1993).

Modeling

Compression Models

Compression models help to understand the behavior of the biomass grinds or particles during pelleting and can help optimize the pressures needed to obtain a quality pellet. Compression tests and models have been widely used for pelletization in the areas of metal, pharmaceuticals, agricultural, and food. Numerous equations expressing the relationship between pressure and time during compaction of different raw materials are available in literature.

A majority of the compression models used in pharmaceutical and biomass materials has been discussed and reviewed in detail by Tabil and Sokhansanj (1996), Adapa et al. (2002a), Denny (2002) and Mani et al. (2003). Kawakita and Ludde (1971) and Mani et al. (2004) suggested that among the different compression models, the Heckel and Cooper-Eaton models are widely used in understanding the compression behavior of pharmaceutical, cellulosic, and soft and fluffy materials. Tabil and Sokhansanj (1996a & b) and Adapa et al. (2002a) studied the applicability of the models for alfalfa pellets and concluded that Cooper-Eaton, Heckel, and Panelli-Filho models provided a better fit to the compression data. The compression models most widely used in biomass densification are described in the following sections (Tabil and Sokhansanj, 1996a & b; Tabil, 1996; Mani et al., 2002 and Adapa et al., 2009).

In his studies on compression and compaction behaviour of fractionated alfalfa grinds, Adapa et al. (2002a) concluded that the Cooper-Eaton model provided the best fit for experimental data obtained from samples at 8-9% moisture content, while the Heckel and Panelli-Filho models provided the best fit for experimental data obtained from samples at 13-14% moisture content (except for those having 75% leaves).

Studies on understanding the compression characteristics of ground agricultural straws like barley, canola, oat and wheat has indicated that the Kawakita-Ludde model provided an excellent fit having an R^2 values of 0.99 (Adapa et al., 2009). The same authors concluded that the parameters of the Cooper-Eaton model best represented the densification of ground straw samples by the particle rearrangement method. Furthermore, the Jones model indicated that canola and oat straws were more compressible compared to barley and wheat straws (Adapa et al., 2009).

Spencer and Heckel Model

The Spencer and Heckel model is used to express density in terms of packing fractions as a function of applied pressure. The following equations are used to describe the compression behavior of powder materials. The constants *m* and *b* describe the two stages of compression: (1) pre-occupation and (2) particle rearrangement due to densification.

$$\ln\frac{1}{1-\rho_f} = m\rho + b \tag{2}$$

where, $$\rho_f = \frac{\rho}{\rho_1 X_1 + \rho 2 X_2} \tag{3}$$

Shivanand and Sprockel (1992) suggested that constant *b* be related to relative density at particle rearrangement (ρ_f) by the following equation:

$$b = \ln \frac{1}{1-\rho_f} \tag{4}$$

Equation (2) indicates that higher ρ_f will result in greater volume reduction due to more particle rearrangement and *m* is the reciprocal of the mean yield pressure required to induce elastic deformation. Higher *m* values of the fitted data will indicate the onset of plastic deformation due to low yield pressures indicating the material is more compressible.

Walker Model

Walker (1923) developed a model based on the experimental data from compressibility of powders and expressed the volume ratio (V_R) as a function of applied pressure (P).

$$V_R = m \ln P + b \tag{5}$$

and

$$V_R = \frac{V}{V_s} \tag{6}$$

where

P = the applied pressure (MPa);

V_R = the volume ratio;

V is the volume of compact at pressure P (m^3), and

V_s *is the* void free solid material (m^3)

Jones Model

Jones (1960) developed a model for the density and pressure data of compacted metal powder.

$$\ln \rho = m \ln P + b \tag{7}$$

where

$$b = \ln(\frac{1}{1-\rho_0})\ and\ \rho f = \frac{\rho}{\rho_1 x_1 + \rho_2 x_2} \tag{8}$$

ρf = the packing fraction or relative density of the material after particle rearrangement;

ρ_o = the relative density of the powder mixture (kg/m^3);

ρ_1 and ρ_2 = the particle density of component of the mixture (kg/m^3);

x_1 and x_2 = mass fraction of the component of the mixture, and

m and *b* = constants determined by the slope and intercept of the extrapolated linear region of the plot.

Higher values of ρf indicate that there will be more volume reduction of the samples due to particle rearrangement. Furthermore, constant *m* is shown as reciprocal to the mean yield pressure required to induce the elastic deformation (York and Pilpel 1973). A large *m* value will indicate that the yield pressure is low and the plastic deformation onsets at relatively low pressures indicate that the material is more compressible.

Cooper-Eaton Model

Cooper Eaton model assumes compression as nearly two independent probabilistic processes: (1) filling of voids having equal size as particles and (2) filling of voids smaller than particles.

$$\frac{V_0 - V}{V_0 - V_s} = a_1 e^{-\frac{k_1}{P}} + a_2 e^{-\frac{k_2}{P}} \tag{9}$$

where

V_o = the volume of compact at zero pressure (m^3), and

a_1, a_2, k_1, k_2 = Cooper-Eaton model constants.

According to Comogly (2007) the practical difficulty in the application of the Cooper-Eaton model is to understand the physical significance of the constants in the equation and realize that it is more suitable to one component system.

Kawakita and Ludde Model

The Kawakita and Ludde model includes the pressure and volume factors.

$$\frac{P}{C} = \frac{1}{ab} + \frac{P}{a} \tag{10}$$

and

$$C = \frac{V_0 - V}{V_0} \tag{11}$$

where

C is the degree of volume reduction or engineering strain, and

a and b are the Kawakita- Ludde model constants related to characteristics of the powder.

The linear relationship between *P/C and P* allows the constants to be evaluated graphically. This compression equation holds true for soft and fluffy powders (Denny 2002; Kawakita and Ludde 1971). Any deviation from this expression is sometimes due to fluctuations in the measured value of V_0. Mani et al. (2004) indicated that constant a is equal to the initial porosity of the sample, while constant $1/b$ is related to the failure stress in the case of piston compression.

Sonnergaard Model (Log-Exp-Equation)

Sonnergaard (2001) model is a log-exp equation that considers two processes simultaneously: (1) a logarithmic decrease in volume by fragmentation, and (2) an exponential decay representing plastic deformation of powders.

$$V = V_1 - w \log P + V_0 \exp(-P/P_m) \tag{12}$$

where

V_1 is the volume at pressure 1 (MPa);

P_m is the mean pressure (MPa), and

w is a constant.

Sonnergaard (2001) suggested that his model provides better regression values compared to Cooper-Eaton model and Kawakita and Ludde model. This model is suitable for medium pressure applications (~ 50 MPa).

Panelli-Filho Model

Panelli-Filho model is given by the following expression:

$$\ln \frac{1}{1-\rho_r} = A\sqrt{P} + B \tag{13}$$

Where

ρ_γ = the relative density of the compact;

A is the parameter related to densification of the compact by particle deformation, and

B is the parameter related to powder density at the start of compression.

Response Surface Models

The process of densification is complex and involves a lot of operational and feedstock variables. Experimental designs, data analysis using statistical and evolutionary methods and further optimization will be of a great advantage to finding the best quality attributes and better understanding the complex system. Shankar and Bandyopadhyay (2004 & 2007) and Shankar et al. (2008) successfully used genetic algorithms and artificial neural networks to understand and optimize an extrusion process. In their studies they used a combination of RSM and GA for a better understanding of the extrusion pelletization process.

Experimental Designs

Experimental designs are widely used for product development with minimum number of experimental runs. These designs help to understand the affect of process variables on the quality of the product. Different experimental designs like factorial, central composite, Box-Behnken, and rotatable can be used to perform the densification experiments in an economical way. According to Mullen and Ennis (1979) rotatable experiments are the best for the product development. The main advantage of rotatable designs over the other mentioned is that it minimizes the number of experiments for a given set of process variables tested. In general, the objective of the researchers or commercial processers is to find the optimal conditions using a small number of level combinations and experiments.

These experimental designs help the researcher perform experiments for each variable at different levels. In general, each variable will have a number of levels (this is a range of values over which the variable will vary). When these levels are combined for all the variables, it gives the total number of level combinations. The levels of the variables are generally divided into a lower average value, center value, and high average value. Other levels are also possible between the lower and center levels. In rotatable and other experimental designs all the variables are coded and the calculations are performed using coded variables, which are converted to the original variables at the conclusion of the analysis.

Many researchers (Shankar and Bandyopadhyay, 2004; Shankar and Bandyopadhyay, 2005; Shankar et al., 2008; Rout and Bandyopadhyay, 1999; and Bandyopadhyay and Rout, 2001) have successfully used these designs for developing response surface models to understand the affect of process variables on product characteristics and create optimization routines.

Response Surface Models

Response surface methodology (RSM) is a collection of mathematical and statistical techniques used for modeling and analyzing problems where the response of the interested variable is influenced by several other variables and the objective is to optimize this response. The general form of the response surface model is given by the following equation (Montgomery, 1976).

$$y = f(x_1, x_2, x_3, \ldots . x_n) + \varepsilon \tag{14}$$

Where

y = the response or dependent variable;

$x_1, x_2, x_3, \ldots . x_n$ = the process or independent variables, and

ε = the noise or error observed in the response variable y.

In general, the first step in RSM modeling is to find a suitable approximation for the true functional relationship between y and the set of independent variables. Usually a low-order polynomial in some region of the independent variables is considered. If the response is well modeled by a linear function of the independent variables, then the approximating function is a first-order model.

The general form of first order model is:

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \ldots\ldots\ldots\ldots \beta_k x_k + \varepsilon \tag{15}$$

If the process involves curvature in the system, then a polynomial of higher degree must be used, such as second-order model.

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^{\,2} + \sum\sum_{i<j} \beta_{ij} x_i x_j + \varepsilon \tag{16}$$

In general, all the RSM models utilize one or more of these models. It is unlikely that a polynomial model will be a reasonable approximation of the true functional relationship over the entire space of the independent variables, but for a relatively small region they usually work quite well. The objectives of these models are to find the optimum operating conditions for a system to determine a region where the requirements are most satisfied (Montgomery, 1976).

Optimization Methods

First Order Models Analysis-Steepest Ascent or Descent Method

The method of the steepest accent is a procedure for moving sequentially along the path of the steepest ascent or in the direction of the maximum increase on the response. In the case of minimization, it is called steepest descent method. The direction of the steepest ascent is the direction in which the response increases steeply and is opposite to the minimization problem. Usually the path of the steepest ascent is the line through the center of the region of interest and normal to the fitted surface, indicating that the steps along the path are proportional to the regression coefficients. Experiments will be conducted along the path of steepest ascent until no further increase in response is observed. This procedure is continued till the experimenter reaches the vicinity of the optimum (Montgomery, 1976).

Second Order Models Analysis

In second order model analysis, the experimenter wants to find the optimum set of operating conditions for the stationary point, or x_s, and to characterize the nature of the response surface (Montgomery, 1976). The stationary point is given by the following equation.

$$x_s = -\frac{1}{2} B^{-1} b \tag{17}$$

A further stationary point is characterized to find out whether it is point of maximum or minimum response or a saddle point. The response can be predicted at a stationary point using the below equation.

$$\hat{y} = \hat{\beta}0 + \frac{1}{2} x_s' b \tag{18}$$

where

$$b = (k \times 1) \tag{19}$$

is the vector of the first-order regression coefficients, and

β is a *(k×k)* symmetric matrix whose main diagonal elements are pure quadratic coefficients $\left(\hat{\beta}_{ii}\right)$ and whose half diagonal elements are one-half of the mixed quadratic coefficients $\hat{\beta}_{ij}, i \neq j$.

Contour Map Analysis

Contour map analysis is more or less a straight forward method if the number of variables are less than two or three. Construction and interpretation of the contour plots are relatively easy. Also, when there are few variables, a more formal analysis called the canonical analysis may be more useful (Montgomery, 1976).

RSM is considered an intuitively simple method, but precise interpretation is tedious when optimizing a function with more than three independent variables at wider experimental range. Also, solving the RSM equations using canonical analysis involves orthogonal rotation of canonical variants where the interpretation is considered very difficult (Hotelling, 1935 &1936). Nie et al. (1970) in their study stated that by using canonical analysis, the user has to examine two sets of coefficients simultaneously, which may often be very complex and difficult to interpret. To overcome these practical problems, advanced computational methods, which are based on evolutionary operations, are now commonly used for optimization of multivariate or complex problems.

Optimization using Evolutionary Algorithms

Genetic algorithm

Living organisms are consummate problem solvers where the organisms come by their abilities through the apparently undirected mechanism of evolution and natural selection. Researchers who search for solutions for complex problems see evolution as a remarkable power that can be emulated. The process of natural selection helps to eliminate one of the greatest hurdles in software design, which is specifying in advance all the features of a problem and the actions that should be taken to deal with the problem. By harnessing the mechanisms of evolution, researchers should be able to “breed” programs that solve problems even when no person can fully understand their structure (Holland, 1992). The process of natural selection, which is a typical feature of genetic algorithms, makes it possible to explore a far greater range of potential solutions to a problem than do conventional programs (Holland, 1992).

Genetic algorithms (GAs) are exhaustive search tools that have gained popularity in process engineering design and new optimization techniques. These tools, based on evolution, have shown remarkable advantages for multiple process parameters optimization. GAs, being a powerful stochastic optimization technique based on evolution methods, find extensive application where process systems are highly complex and nonlinear (Holland, 1992; Gen and Cheng, 1997; Goldberg, 2001; Deb, 2000; Chen and Ramaswamy, 2002, Shankar et al., 2008 and Shankar and Bandyopadhyay, 2004). By harnessing the mechanisms of evolution, GAs makes it possible to explore a greater number of potential solutions to a problem than can be done with conventional programs and will help “breed” new solutions to complex nonlinear problems (Holland, 1992). The main advantages of GA when compared to other gradient-based approaches are the ability to (1) explore the search space more thoroughly with a smaller number performances evaluations than those based on local search, such as simulated annealing, and (2) be less dependent on the good selection of starting points that do not require neighborhood definition (April et al., 2003). Fig. 11 is a flow diagram for GA-based algorithms. The flow diagram of the experimental design and further analysis of the data using statistical and evolutionary algorithms that helped in understanding the complex densification process is given in Fig. 12.

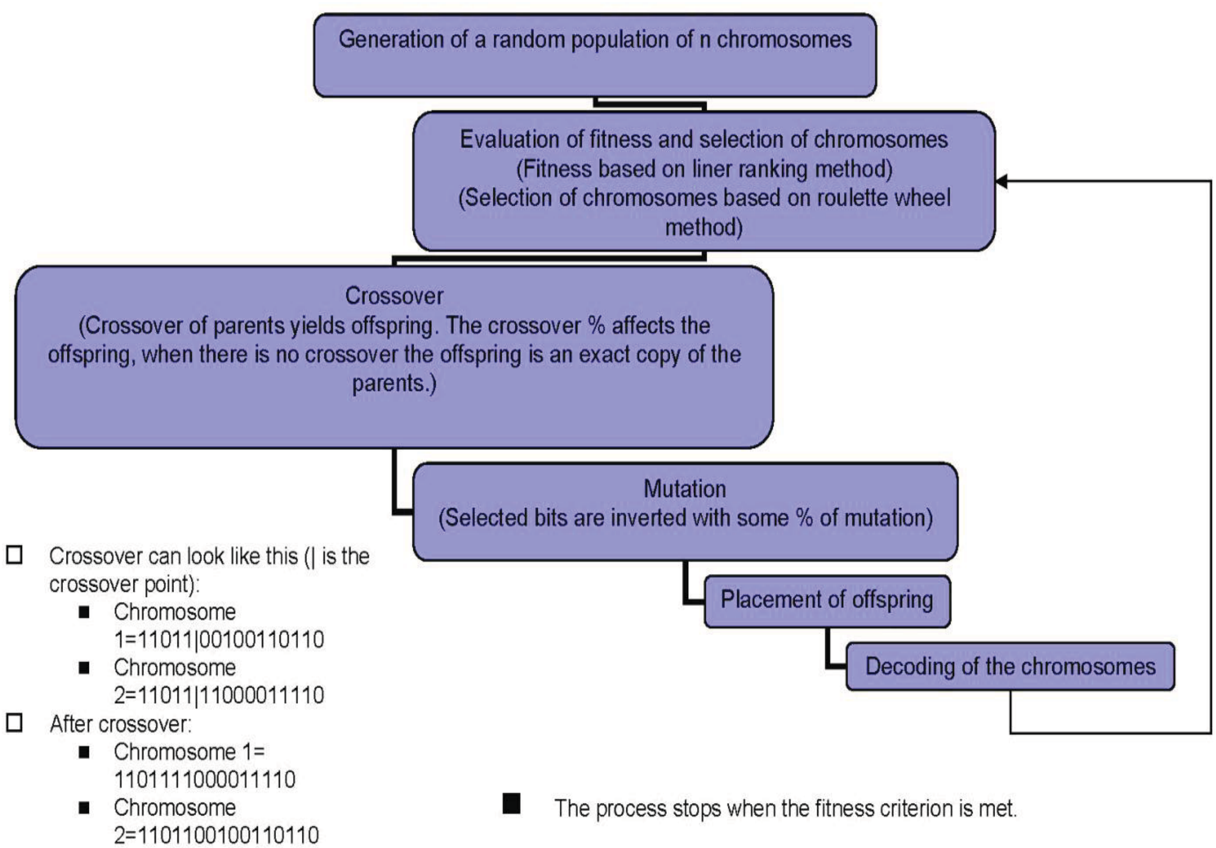


Figure 11. Flow diagram for GA-based algorithm for optimization (Tumuluru et al., 2010)

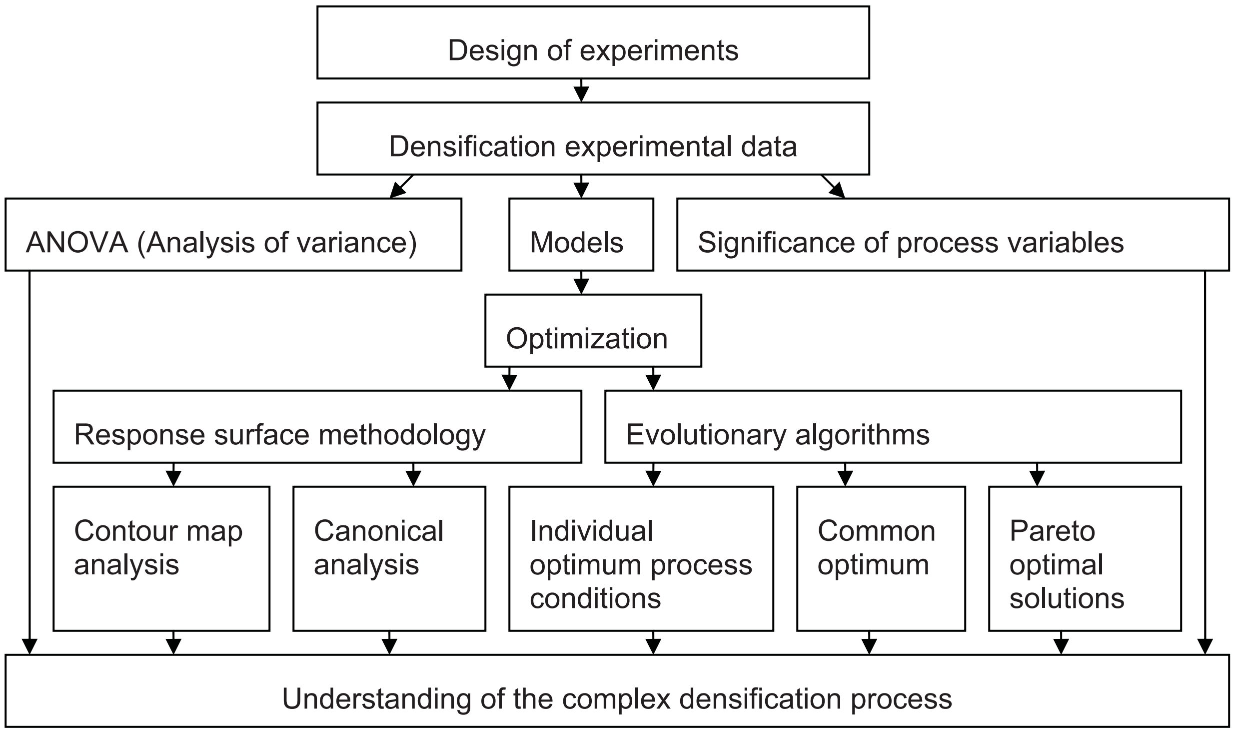


Figure 12. Flow diagram of the densification experimental data analysis using combination of response surface methodology and evolutionary algorithms.

Conclusions

There are many performance parameters and biomass variables that influence the densification process and resulting product characteristics. Understanding these parameters and variables is critical for controlling the cost and performance of the densification process and producing a quality densified product. This paper has identified several densification processes and the mechanisms by which they function in order to provide a better understanding of this much needed operation for bioenergy. A few key results of this review are as follows:

1. The three stages of densification include
   a. Rearrangement of the of particles to form closely packed mass
   b. Plastic and elastic deformation of the particles and
   c. Significant reduction in volume until the density of the pellet reaches the true density of the component ingredients.
2. Biomass densification is a result of solid bridges forming between biomass particles that are dependent on applied pressures and moisture in the biomass.
3. Screw extruders produce denser biomass materials and have more operational advantages compared to piston presses.
4. The production capacity of pellet mills is not dependent on the density of the raw materials as in the case of piston and screw presses.
5. The process variables like temperature, pressure, retention time and relaxation time play a major role on the quality attributes like durability and density of densified biomass.

6. Feedstock variables like moisture content and particle size and distribution play a major role in the flow behavior of raw biomass material through any densification system.
7. Biomass compositions like starch, fat, and lignin influence the quality attributes and behavior of biomass material during the densification process.
8. Preprocessing biomass by grinding, steam conditioning or explosion, or torrefaction significantly improve the binding behavior of the material during the densification process.
9. The addition of binders like protein and starch can improve the binding characteristics of the material during densification.
10. The Kawakita-Ludde model best describes the compression characteristics of agricultural biomass samples.
11. Data analysis using both statistical methods like ANOVA and response surface methods and evolutionary algorithms can help to better understand the complex densification process.

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