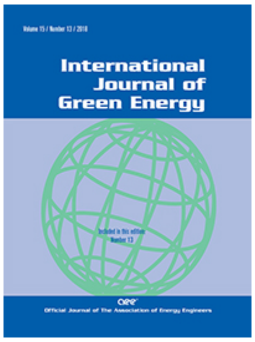

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International Journal of Green Energy

ISSN: 1543-5075 (Print) 1543-5083 (Online) Journal homepage: www.tandfonline.com/journals/ljge20

Renewable biofuel production from biomass: a review for biomass pelletization, characterization, and thermal conversion techniques

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INTERNATIONAL JOURNAL OF GREEN ENERGY
2018, VOL. 15, NO. 13, 837–863
https://doi.org/10.1080/15435075.2018.1529581

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Renewable biofuel production from biomass: a review for biomass pelletization, characterization, and thermal conversion techniques

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ABSTRACT

The exhaustion of fossil fuel resources has instigated a necessity to find new alternatives like biofuels, for heat and power generation. Biofuels are usually generated from the thermal conversion of densified biomass material. Densification systems convert biomass into pellets, and consist of three phases: pre-pelletization, pelletization, and post- pelletization.

This article provides an overview of available biomass densification techniques. A detailed discussion has been provided to emphasize the effect of raw material properties on the pellet's durability and bulk density. A quality parameter (Q) has been proposed to evaluate the quality of pellets considering the factors involved in pellet characterization. Particularly, accounting for pellet compression rather than tensile forces were found to be better when quantifying the pellet quality parameter Q.

A discussion regarding the binding mechanisms, types of binders used, and their effect on the pellet's durability is provided, in addition to the pelletization process by itself with the main parameters that affect its operation. The post-pelletization processes were presented, focusing on three thermal conversion techniques: gasification, pyrolysis, and combustion. A comparison between these techniques has been provided, in addition to recommendations regarding pros and cons of each one. Finally, the environmental footprints of densification systems have been reviewed.

ARTICLE HISTORY
Received 15 November 2017
Accepted 25 September 2018

KEYWORDS
Biomass; pelletization; thermal conversion; binders; life cycle assessment

1. Introduction

The increasing global energy consumption, coupled with escalating fuel prices, has prompted the need to look for alternative energy sources. The current trend is to employ biofuels, produced from the thermal conversion of biomass waste material, which renders such fuels as environmentally friendly options that aim to produce energy from waste. A rapid growth has been reported in terms of the production and consumption of wood biomass waste material, especially for heating and power production activities, throughout the past couple of years. According to the European Bioenergy Outlook (European Bioenergy Outlook (AEBIOM) 2013), the world's wood pellet production has been reported to be around 24.5 Mton, with a total of 50% produced in the European countries. However, to date, biomass has not been utilized extensively as an energy source because of its moderately low energy density when related to other energy sources, such as fossil fuels, and natural gas. Their low energy density often causes costly transportation, and introduces many difficulties in storage and handling. To overcome these problems, biomass can be densified into a form that can be stored and handled using similar techniques that many grain industries adopt for their handling and storage (Fasina and Sokhansanj 1996, 1992; Sokhansanj and Wood 1991). One form of densified biomass can be attained using pellets. Biomass pelletization involves increasing the bulk (or apparent) density of the material to create a denser and more durable form (Mani and Chandra. 2003). This considerably reduces transportation, handling, and storage costs (Holm et al. 2006; Nilsson, Bernesson, and Hansson 2011; Werther et al. 2000).

Since pellet quality is largely affected by the type of feedstock and its properties, it is very important to discuss how raw material composition, moisture content, and particle size distribution, affect the pellet's performance. For instance, a homogeneous pellet size and shape (Theerarattananoon, Xu, and Wilson 2011) is often desirable, since this prevents the blocking and obstruction of biomass livestock feeders, especially those used for thermal applications such as domestic heating. Moreover, some types of biomass feedstock require the addition of binders, in order to enhance the intermolecular bonding within the pellet, so as to avoid failure during transportation. However, it should be noted that some of the physical and thermal parameters of wood pellets are often influenced by the usage of binding agents. According to the EU standards, binders that enhance pellet's quality, reduce emissions, or improve the burning efficiency of the pellets can compose only 2% of the pellet's total mass (European Pellet Council 2013; Tarasov, Shahi, and Leitch 2013). The most recognized kinds of pellet binders are lignosulphonate, starch, dolomite, corn/potato flour, and some forms vegetable oils (Pellet Fuel Institute 2010; Tarasov, Shahi, and Leitch 2013). Different binding agents affect manufacture costs differently; hence, a lot of attention must be given to the type of binding agents used for pelletization.

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Pellet durability, as well as their bulk density, are the main criteria used to assess the quality of produced pellets. Moreover, assessing the equipment's die parameters such as the hole depth, hole pitch, hole width, and die material, as well as the optimum pressure and temperature conditions for pelletization, is essential. Different pressure conditions will result in different binding processes. At high pressures, the naturally existing binding elements in biomass materials (such as lignin) are pressed outside the atoms, which in turn induce inter-particle bonding (Kaliyan and Morey, 2009a). Pellet dimensions also affect their durability; Heffner and Pfost (1973), concluded that smaller diameters resulted in greater pellet durability. Hence, this review paper discusses the different parameters that affect pellet durability, and their quality in general.

Biomass raw material must be readily available, and combustible in pellet form, to be considered suitable for biofuel production. Therefore, following raw material characterization, it is of great importance to study the pellet performance to determine suitable raw material options for biofuel production during later stages. This is usually accomplished by subjecting the pelletized form of the respective biomass material through various fuel burning techniques. Therefore, this review paper compares three different modes of pellet utilization: gasification, pyrolysis, and combustion (also known as oxidation), mainly in terms of the quantity of oxygen gas needed for burning, the temperature required for the process, as well as the products/byproducts obtained. In order to enable the selection of clean pellet-thermal conversion techniques that ideally produce zero waste, it is important to investigate key parameters that affect each process, quantify and assess the products and byproducts obtained from each, and propose possible ways for product/byproduct utilization.

Hence, this review paper is organized as follows: the pre-pelletization process will be first introduced, where the types of the biomass raw material will be discussed, followed by the biomass elemental composition, taking into consideration the raw material moisture content and particle size distribution, and their effect on the pellets durability and bulk density. Then, the main binding mechanisms will be presented, in addition to the types of the binders used and their effect on the pellets properties. After that, the pelletization process will be presented, discussing the affecting parameters in the pelletization process and how they affect the final quality of the produced pellets. The post- pelletization processes are then introduced regarding the different thermal conversion models and the main product and mechanisms. The paper finally sums up with introducing the Life Cycle Analysis, LCA, method for assessing the environmental impacts of the whole process with presenting a summary from literature.

2. Pre- pelletization process

In order to guarantee that durable pellets with suitable bulk density specifications are obtained, the biomass raw material should first be treated and marginally modified through a pre-pelletization stage. Therefore, properties like chemical composition, moisture content and particle size distribution of the raw material feed are very important, and greatly affect the pellet durability, and the bulk density of the pellets attained.

3. Types of biomass raw material

Numerous types of biomass raw materials have been evaluated to produce high-quality pellets. For example, pellets from tomato residues were investigated by Celma, Cuadros, and López-Rodríguez (2012). Pellets from grape pomace and pyrenean oak were experimented by Miranda et al. (2011). Different olive pruning residues were studied and characterized by Garcia-Maraver et al. (2015) in which olive leaves (OL), olive pruning (OP) and olive wood (OW) were classified as 3 different samples. The physio-chemical characteristics of pellets made up from coppice of short rotation tropical hardwood (Gmelina arborea, Acacia mangium, and Paraserianthes falcataria) were studied by Acda (2015), whereas spruce wood sawdust, as biomass waste, is also considered a candidate as a renewable fuel, however its bulkiness, low energy density, high volatiles content, and NOx emission are problematic. Another study was also conducted by Fasina (2008) where the physical properties of peanut pellets were compared to values of switchgrass (McLaughlin et al. 1999; McLaughlin and Kszos 2005). Even poultry litter was investigated by McMullen et al. (2005) as a potential raw material for biofuels. Other examples of biomass raw material, which may be used for pellet production include: soy bean straw (Huang et al. 2016), alfalfa stems (Boateng et al. 2008), spruce and beech (Rath et al. 2002), and sugarcane bagasse (Erlich et al. 2005; Yin 2012). To sum up, Table 1 presents a summary of the most common biomass raw materials, found in literature, that are used for pellet making.

4. Biomass composition and characterization

Generally speaking, the components of biomass raw material are usually studied through ultimate and proximate analysis techniques. Proximate analysis involves measuring the values of the moisture content (MC), ash content, volatile matter contents (VM), and fixed carbon contents (FC) of the raw material. On the other hand, ultimate analysis measures carbon, hydrogen, sulfur, nitrogen, and oxygen elements. Since both analysis techniques are able to unveil the main elements that are present in biomass raw material, the corresponding physical and chemical properties of produced pellets from a given raw material may be easily predicted, once the main raw material elements are measured.

Yin (2012) conducted both ultimate and proximate analysis on wheat straw, cotton, rice straw bamboo, stalk, corn stover, hybrid poplar, and oak wood, to predict the respective heating values for each different raw material. Serrano et al. (2011) studied the effect of moisture content, particle size, and pine addition on the quality of pellets made from barley straw. (Biswas, Yang, and Blasiak 2011) investigated steam explosion pretreatment options to discrete the main elements of biomass made from wood to promote the pellet production. The main findings from the ultimate and proximate analysis of various types of raw materials are summarized in Figures 1 and 2, respectively.

Following the various results reported from studies that performed ultimate analysis for biomass feed characterization (Figure 1), it is observed that the main component of biomass

Table 1. Summary of raw materials used for pellet production (modified from: Stelte et al. 2011).

Biomass Resource	Reference
Pine	Arshadi et al.,2013; Filbakk, 2011; Finell et al. 2009; Holm et al. 2007; Li et al. 2012; Li and Liu 2000; Nielsenet al. 2009a, 2009b, Nielsen, Gardner, and Felby 2010; R. Samuelsson, 2009; Arrieche et al. 2011
Spruce	Arshadi et al., 2013; Finell et al. 2009; Holm et al. 2006, Holm et al. 2007; Rhen et al. 2005; Samuelsson et al., 2009; Stelte et al. 2012; Stelte et al. 2011d; Stelte et al., 2011a
Beech	Holm, 2006, 2007; Stelte et al. 2011d; Stelte et al. 2011a
Eucalyptus (EUC)	Pirraglia, 2010, 2012; Telmo and Lousada 2011
Poplar	Nielsen et al. 2009b
Oak (OAK)	Li et al. 2012; Nielsen, 2010; Castellano et al. 2015
Salix	Biswas et al. 2011
Fir	Lam et al. 2011
Alfalfa	Wongsiriamnuay PK, 2002; Tabil 1996; Puig-Arnavat et al. 2016
Barley	Faborode, 1987; Mani, 2004; Mani, 2003; O'Dogherty, 1984; Serrano et al. 2011
Canola	Adapa, 2009; Tilay et al. 2015; Chico-Santamarta et al. 2012
Oat (OAT)	Adapa, 2009
Wheat	Sokhansanj, 1992; Mani, 2004; Mani,2003; O'Dogherty, 1984; Ollet, 1993; Stelte et al. 2012; Stelte et al. 2011d; Stelte et al. 2011a, b; Theerarattananoon, Xu, and Wilson 2011; Li and Liu 2000
Rice Straw (RIC)	Chen, 2011; Faborode, 1987; Said et al. 2015; Ishii and Furuichi 2014; Castellano et al. 2015
Rye	Narra et al. 2010
Reed Canary Grass	Larsson et al., 2008
Corn Stover	Kaliyan et al. 2009c; Kaliyan and Morey 2009b; Li et al. 2012; Theerarattananoon, Xu, and Wilson 2011; Mani, 2004; Mani, 2003; Tumuluru 2016
Corn Cobs	Kaliyan and Morey 2010a
Soy Bean	Chen, 2011; Xin Huang et al. 2016
Switch Grass	Gilbert, 2009; Kaliyan and Morey 2009b, Kaliyan et al. 2009c; Kaliyan and Morey 2010; Mani, 2004; Mani, 2003
Big Blue Stem	Theerarattananoon, Xu, and Wilson 2011
Sugar cane Bagasse	Erlich et al. 2005
Cotton	Coates 2000; Li and Liu 2000
Olive (Olive leaves (OL), olive pruning (OP) and olive wood (OW))	Choi, 2009; Barbanera et al. 2016; Garcia-Maraver et al. 2015; Christoforou, Kylili, and Fokaides 2016
Peanut Hulls	OO, 2008
Mixed Residues	Gil et al. 2010; Harun and Afzal 2016
Tomato Residues	Ruiz Celma, 2012; Cuadros, and López-Rodríguez, 2012
Grape Pomace	Miranda et al. 2011
Poultry Litter	McMullen, 2005
Agricultural Biomass	Ignacy Niedziołka, 2015; Karkania, Fanara, and Zabaniotou 2012
Wood Residues	Oveisi et al. 2013; Graham M. H., 2002; Sgarbossa et al. 2014; Vaughn et al. 2013; Xin Huang et al. 2016; Amit Kumar Biswas, 2011; Nguyen et al. 2015; Lu et al. 2014; Alakoski et al. 2016; Yazdanpanah 2009; Mola-Yudego, 2014; Tumuluru 2016
Maize Residue	Wongsiriamnuay PK, 2002
Bamboo	Liu et al., 2010
Short Rotation Tropical Hardwood (Gmelina arborea (G. arborea), Acacia mangium (A. mangium) and Paraserianthes falcataria (P. falcataria))	Acda 2015

raw material is carbon, with an average value of. 46.87 ± 0.74% and a standard deviation of 3.79%. (Cuadros et al. 2011; M.T. Miranda et al. 2011; Choi, Kim, and Cha 2009; M.Barbanera et al. 2016; A. Garcia-Maraver et al. 2015; E. Christoforou, Kylili, and Fokaides 2016; Fasina 2008; McMullen et al. 2005; Adapa et al. 2002; M. Puig-Arnavat et al. 2016; Arshadi et al. 2008; Stelte et al. 2011d; Erlich et al. 2005; Adapa, Tabil, and Schoenau 2009; Mani et al. 2006; Liu et al. 2016; Kaliyan and Morey 2009a, 2009b; Chen et al. 2011; Nielsen, Gardner, and Felby 2010b; Mani, Tabil, and Sokhansanj 2004; Biswas, Yang, and Blasiak 2011; Chen et al. 2011).

Nitrogen and sulfur are usually present in relatively small quantities, with average values of 1.14 ± 0.18% and standard deviation of 0.93% for nitrogen, and an average of 0.23 ± 0.18% and standard deviation of 0.91% for sulfur (A. Ruiz Celma et al. 2011; M.T. Miranda et al. 2011; Choi, Kim, and Cha 2009; M.Barbanera et al. 2016; A. Garcia-Maraver et al. 2015; E. Christoforou, Kylili, and Fokaides 2016; Fasina 2008; McMullen et al. 2005; Adapa et al. 2002; M. Puig-Arnavat et al. 2016; Arshadi et al. 2008; Stelte et al. 2011d; Erlich et al. 2005; Adapa, Tabil, and Schoenau 2009; Mani et al. 2006; Liu et al. 2016; Kaliyan and Morey 2009a, 2009b; Chen et al. 2011; Nielsen, Gardner, and Felby 2010b; Mani, Tabil, and Sokhansanj 2004; Biswas, Yang, and Blasiak 2011; Chen et al. 2011). It worth noting that problems of nitrogen oxide emissions could arise in raw materials with high nitrogen levels, especially materials that contain more than 2% (wt) nitrogen. Moreover, it is essential to measure the amount of sulfur in the raw matter in order to be able to estimate the amount of sulfur gas release during thermal conversion. Energy industries always find biomass raw materials with low levels of nitrogen and sulfur extremely desirable for biofuel production, since they are classified as more environmentally friendly and less corrosive.

On the other hand, results attained from studies that conducted proximate analysis for biomass feed characterization are summarized in Figure 2. The total amount of volatile matter, ash, and fixed carbon content, were reported to be quite different for different raw materials. Generally speaking, volatile matter content was the highest, with an average value of 76.68 ± 1.69% and standard

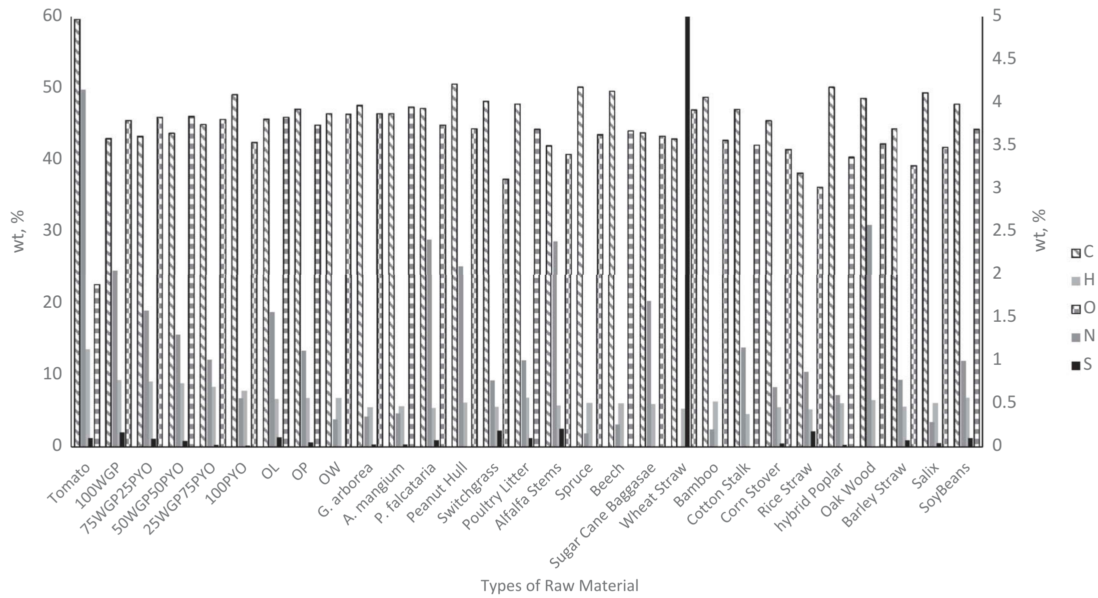

Figure 1. Ultimate Analysis for Different Types of Raw Material (Generated from: Mani et al., 2003; Acda 2015; Erlich et al. 2005; Fasina 2008; Garcia-Maraver et al. 2015; Holm et al. 2007; Li et al. 2012; Mani, Tabil, and Sokhansanj 2004; McMullen et al. 2005; Miranda et al. 2011; Theerarattananoon, Xu, and Wilson 2011).

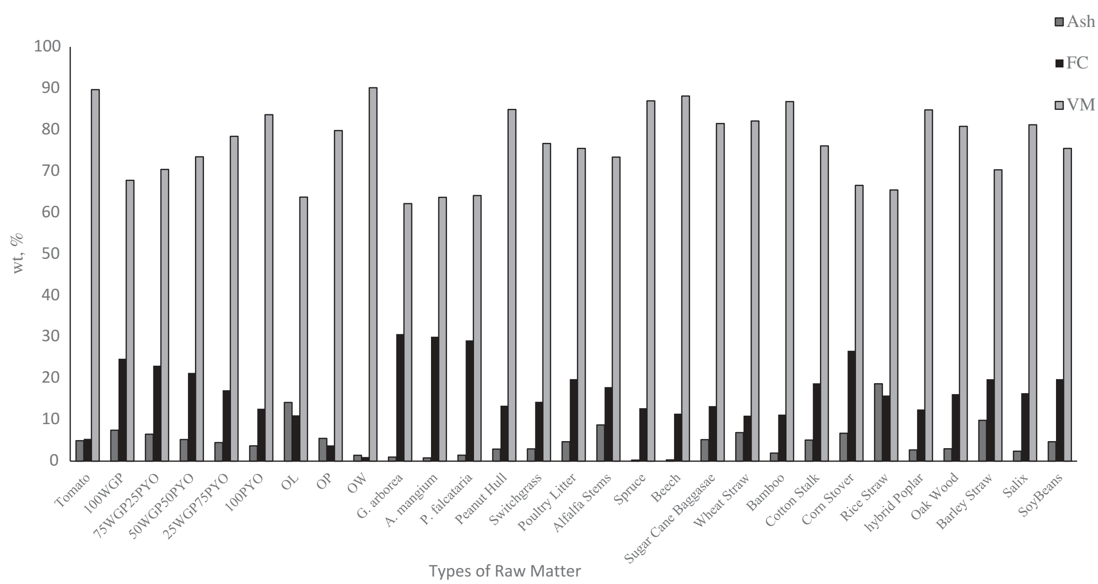

Figure 2. Proximate Analysis for Different Types of Raw Material (Generated from: Mani et al., 2003; Theerarattananoon, Xu, and Wilson 2011; Celma, Cuadros, and López-Rodríguez 2012; Miranda et al. 2011; Garcia-Maraver et al. 2015; Acda 2015; McMullen et al. 2005; Fasina 2008; Erlich et al. 2005; Holm et al. 2007; Li et al. 2012; Li and Liu 2000; Nielsen et al., 2009a, 2009b, 2010; Castellano et al. 2015; Biswas, Yang, and Blasiak 2011; Puig-Arnavat et al. 2016; Chen et al. 2011; Kaliyan and Morey, 2009b).

deviation of 8.6% (Cuadros et al. 2011; M.T. Miranda et al. 2011; Choi, Kim, and Cha 2009; M.Barbanera et al. 2016; A. Garcia-Maraver et al. 2015; E. Christoforou, Kylili, and Fokaides 2016; Fasina 2008; McMullen et al. 2005; Adapa et al. 2002; M. Puig-Arnavat et al. 2016; Arshadi et al. 2008; Stelte et al. 2011d; Erlich et al. 2005; Adapa, Tabil, and Schoenau 2009; Mani et al. 2006; Liu et al. 2016; Kaliyan and Morey 2009a, 2009b; Chen et al. 2011; Nielsen, Gardner, and Felby 2010b; Mani, Tabil, and Sokhansanj 2004; Biswas, Yang, and Blasiak 2011; Chen et al. 2011), while ash content was the lowest in many of the cases, with average values of 4.95 ± 0.79% and standard deviation of 4.06%. Moreover, the ash content will vary significantly, depending on the type of biomass raw material. For instance, biomass derived from spruce and beech were found to be associated with ash contents that are below 0.5%, while biomass derived from rice straw are often associated with higher ash content values, typically 18% and above. (Cuadros et al. 2011; M.T. Miranda et al. 2011; Choi, Kim, and Cha 2009; M.Barbanera et al. 2016; A. Garcia-Maraver et al. 2015; E. Christoforou, Kylili, and Fokaides 2016; Fasina 2008; McMullen et al. 2005; Adapa et al. 2002; M. Puig-Arnavat et al. 2015; Arshadi et al. 2008; Stelte et al. 2011d; Erlich et al. 2005; Adapa, Tabil, and Schoenau 2009; Mani et al. 2006; Liu et al. 2016; Kaliyan and Morey 2009a, 2009b; Chen et al. 2011; Nielsen et al. 2012; Mani, Tabil, and Sokhansanj 2004; Biswas, Yang, and Blasiak 2011; Chen et al. 2011). Volatile matter includes all components that are released in the absence of oxygen under high temperature conditions, except for water. Volatile matter is a key health and safety concern, since materials high in volatiles have an increased risk of spontaneous combustion. Agricultural residues often contain high amounts of volatile matter, making them easier to catch fire and burn. Different values for ash content are obtained for different raw materials, due to variances composition. Raw materials with a high ash content are often less favorable, due to the various problems that arise during ash removal, and the need for frequent cleaning and maintenance of the equipment. The fixed carbon content is calculated by subtracting the total moisture, volatile matter, and ash content from the original mass of the raw material sample, where average values of ash were found to be 16.59 ± 1.44% and standard deviation of 7.38% (Cuadros et al. 2011; M.T. Miranda et al. 2011; Choi, Kim, and Cha 2009; M.Barbanera et al. 2016; A. Garcia-Maraver et al. 2015; E. Christoforou, Kylili, and Fokaides 2016; Fasina 2008; McMullen et al. 2005; Adapa et al. 2002; M. Puig-Arnavat et al. 2015; Arshadi et al. 2008; Holm et al. 2011; Erlich et al. 2005; Adapa, Tabil, and Schoenau 2009; Mani et al. 2006; Liu et al. 2016; Kaliyan and Morey 2009a, 2009b; Chen et al. 2011; Nielsen, Gardner, and Felby 2010b; Mani, Tabil, and Sokhansanj 2004; Biswas, Yang, and Blasiak 2011; Chen et al. 2011). Moisture content of biomass raw material is discussed in the following section.

4.1. Moisture content

Moisture is one of the essential elements in the pellet formation process, since water induces many of the intermolecular forces that are necessary for effective pelletization. However, once the moisture content of biomass raw material exceeds a certain threshold, the resulting pellet quality will decrease, as they become highly prone to degradation. From the proximate analysis in Figure 2, where average values of moisture content were found to be 10.34 ± 0.56% and standard deviation of 3.33%, (Chen et al. 2011; Faborode and Ocallaghan 1987; Niedziołka and Kachel 2015; V. Karkania, Fanara, and Zabaniotou 2012; Wongsiriamnuay et al. 2013; Adapa, Tabil, and Schoenau 2009; Mani et al. 2006; Z. Liu et al. 2016, 2009; A. Tilay et al. 2015; L. Chico-Santamarta et al. 2012; Adapa et al. 2002; M. Puig-Arnavat et al. 2016; Chen et al. 2011; Fasina 2008; McMullen et al. 2005; Pirraglia et al. 2012; Pirraglia, Gonzalez, and Saloni 2010; Telmo et al. 2011; Arshadi et al. 2008; Filbakk et al. 2011; Finell et al. 2009; S. Mani, Tabil, and Sokhansanj 2004; Choi, Kim, and Cha 2009; M.Barbanera et al. 2016; Cuadros et al. 2011), and it can be noted that the moisture content of olive leaves, pruning, and wood studied by Garcia-Maraver et al. (2015), ranged between 7.42% and 11.02%. The grape pomace (GP) and pyrenean oak (PYO) moisture content investigated by Miranda et al. (2011), varied between 7.08% and 7.49%. Niedziołka and Kachel (2015), were able to generate pellets using wheat straw, rape straw, and maize straw biomass, with a moisture content varying between 11.1% and 14.8%. Fasina (2008) analyzed the physical properties of both peanut hull and switch-grass pellets and reported their moisture content to be around 9.94%, and 8.6%, respectively. Colley (2006) studied the effects of moisture content on the physical characteristics of pellets made from switch-grass, using biomass feed with moisture content varying between 9.8% and 15.6%. Carroll and Finnan (2012), measured the moisture content of energy crops and cereals straws, and reported values that are ranging between 9.39% and 13.46%. Castellano et al. (2015), investigated the influence of raw material content, as well as pelletization settings, on the properties of pellets made from different woody biomass such as pine and oak, and non-woody biomass such as oats and rice straw.

It is essential to use biomass raw materials with optimal moisture content values, to achieve better pellet durability, strength, and thermal conversion performance. Absorbed water greatly affects the strengthening of the binding force between pellet particles. Biomass moisture also acts as a lubricant that assists in the formation of van der Waals' forces (Kaliyan and Morey, 2008) by raising the total area of contact between pellet particles.

Extremely high moisture content levels could cause pellet degradation as a result of moisture absorption, since the coupling power drawing the particles together turns out to be not as much as the power pulling the particles separated, as a result of pellet expansion. In contrast, low moisture content causes no significant binding to take effect. This is due to the absence of capillary forces that are responsible for maintaining the pellet structure; hence the pellets become susceptible to breakage. Moreover, it should be noticed that a significant decrease in the moisture content of pellets is witnessed compared to that of the raw material after pelletization, according to the study by Celma, Cuadros, and López-Rodríguez (2012).

To sum up, and according to literature, the maximum moisture content level studied in literature was 14.2%, and the optimum recommended value was reported to range between 8% and 12%. Therefore, having the moisture content of biomass raw material within these optimum values strengthens the binding forces between individual pellet particles. Nonetheless, moisture content levels that are beyond this optimum range will result in severe swelling and pellet disintegration.

4.2. Particle size distribution

Particle size is among the most significant factors that influence the overall pellet quality. In general, greater pellet strength and durability are attributed to the presence of finer particle sizes (MacBain 1966). Fines serve to fill the pore space created by larger particles and promote the development of inter-particles bonds. For this reason, it is essential to pay attention not only to the particle size, but also to the particle size distribution. The ideal pellet quality is attained when a blend of particle sizes is generated, due to the fact that the inter-particle connection, especially mechanical meshing, increases during pelletization. At the same time, inter particle spaces, both attractive and cohesive forces, are eliminated during the process (Kaliyan and Morey, 2009a, 2009b; Payne 1978, 2006).

Generally speaking, as the particle size decreases, the pellet durability increases (Kaliyan and Morey, 2009a), and average particles size was found to be 1.5 ± 0.31 mm, with s standard deviation value of 1.27 mm (Cuadros et al. 2011; M.T. Miranda et al. 2011; Choi, Kim, and Cha 2009; M.Barbanera et al. 2016; A. Garcia-Maraver et al. 2015; E. Christoforou, Kylili, and Fokaides 2016; Fasina 2008; Holm et al. 2011d; Adapa, Tabil, and Schoenau 2009; Mani et al. 2006; Z. Liu et al. 2016; Kaliyan and Morey 2009a, 2009b; Serrano et al. 2011; Arshadi et al. 2008; Lam et al. 2011). Many studies have investigated the effect of particle size on the compaction properties of biomass (Mani et al., 2003; Stelte et al. 2011a, b; Whittaker and Shield 2017). For instance, O'Connell et al. (1995) suggested that a particle size range of 0.6–0.8 mm is recommended for obtaining good quality pellets which is very close to the 0.5–0.7 mm range suggested by Theerarattananoon, Xu, and Wilson (2011). Nonetheless, according to Payne (1978), reasonably good quality pellets can still be derived from different particle size values.

The influence of particle size on a variety of other parameters has also been studied. For example, Stelte et al. (2012) proved that the friction force in the press passage of a pellet mill is in reverse relation with the size of beech particles. This is related to that the contact surface area between the particles and the channel ways increases. Kaliyan and Morey (2009b); (2009c); (2010) proved that a decrease in the particle size for corn stover grinds enhances the density of the produced pellets. On the other hand, a study made by Serrano et al. (2011) found that smaller particles resulted in less dense pellets. Mani, Tabil, and Sokhansanj (2004) found that particle size considerably affects the density of pellets made from barley straw, corn stover, and switch grass. However, Mani, Tabil, and Sokhansanj (2004) reported that this effect was not observed in pellets that were made from wheat straw. It should be noted that the study by Serrano et al. (2011) utilized pelletization press units on an industrial scale while other studies (Mani et al., 2003; Mani, Tabil, and Sokhansanj 2004; Kaliyan and Morey, 2009a, 2009b) involved laboratory scale single pellet press units.

The densification process by itself is very important in determining the optimum particle size. In general, the desired pellet quality depends on a broad variation of particle sizes. For efficient pelletization, particle's diameter should be less than 5 mm. Nevertheless, when the quantity of fines (defined as particles of diameter less than 0.5 mm) is too high in the raw material, this would usually result in a negative effect on friction and pellet quality. Thus, unless binding means are added, the quantity of fines should be in the range 10% to 20%. Jensen, Temmerman, and Westborg (2011) devised a technique that can detect the particle size distribution in a biomass pellet that differs from the one of the raw material before pelletization, since particle size drops during pelletization. It is suggested that wet fragmentation along with mechanical power could be considered as the most convenient technique to identify the inner particle size distribution of a biomass pellet. Thus, many of the studies found in the literature recommend an optimal particle size of less than 5 mm. However, this cannot be considered as a definite rule, since many other factors are involved in the production process of high-quality pellets.

5. Pellet durability and bulk density

Durability is a major factor that is used to describe the physical quality of the pellets. It can be defined as the pellet's capability to tolerate vibrations and shock. According to Colley (2006), pellet durability is considered high if its value exceeds 80%, while it is considered medium if it lies between 70% and 80%, and low if it does not exceed 70%. Durability of pellets can be influenced by different factors, as it will be discussed in the following sections. On the other hand, bulk density is defined as the mass per unit volume of biomass. Pellets with high bulk density values occupy less space, and are easier and cheaper to transport or store. Thus, it is essential to produce pellets with high bulk density values, to be able to enhance transportation and storage efficiency, while reducing incurred costs. Conferring to European standards (European Pellet Council 2013), the bulk density of pellets should be greater than 500 kg/m^3.

5.1. Effect of moisture content on pellet durability

Moisture content greatly affects pellet durability levels. Generally speaking, durability is enhanced when increasing the moisture content up to an optimum. After that, pellet durability starts to decrease after any subsequent increase in moisture contents, beyond the optimum moisture level (Kaliyan and Morey, 2009a, 2009b). For instance, the highest durability levels of pellets made from wheat straw, and corn stover were about 96.8% and stayed constant under moisture content conditions that ranged between 9% and 14%. Pellet durability started decreasing, upon increasing the moisture content above 14% (Theerarattananoon, Xu, and Wilson 2011). Fasina's (2008) study on the physical properties of peanut hull pellets showed that the durability of the peanut hull pellets rose at first to reach a maximum of 90.3% at a moisture level of 9.1%. Increasing the moisture content beyond 9.1% resulted in a decreased pellet durability. It should be noted that durability values are also attributed to many other factors, besides moisture content, such as the particle size, type of pretreatment processing used for the raw material, etc.

The primary increase in durability upon increasing the moisture content is a result of the presence of binding forces from water particles that strengthen the ties between each pellet particles. Any additional increase in moisture levels beyond optimum values causes disruption of the particulate bonds, which triggers the swelling and disintegration of pellets (Fasina 2008; Fasina and Sokhansanj 1996; Singh 2004). Any volume expansion of the pellets, as a result of moisture absorption, is the main cause of pellet swelling. Pellet disintegration then happens as a result of pellet swelling, in which the forces that pull the particles apart become greater than the binding forces that bring the particles together, as in the case of alfalfa pellets (Fasina and Sokhansanj 1992), poultry litter pellets (McMullen et al. 2005) and switchgrass pellets (Colley 2006).

5.2. Effect of moisture content on bulk density

Raw material moisture content greatly affects the attained bulk density values of pelletized biomass. Less dense pellets are usually produced when biomass raw materials with high moisture levels are used, since pellet volume increases with increasing moisture content as a result of pellet expansion. In fact, the source of biomass raw material and its composition significantly affect the pellet bulk density attained.

Colley (2006) investigated the effects of moisture content on the physical properties of pellets from switch-grass. In doing so, they discovered that the switch-grass pellets bulk density dropped as the pellet moisture content increased.

Even though it is not the dominant trend, but generally speaking, pellets with low bulk density values are associated with raw materials with relatively high moisture content. For example, biomass raw materials such as poultry litter, peanuts, corn, and wheat, show a reduction in the pellet bulk density attained with increasing moisture content levels.

There may be several reasons behind this, such as the particle size distribution and raw material pretreatment methods. Serrano et al. (2011) provide several reasons for such a trend, one of which is that moisture content hinders the compaction process. Moreover, the drop in the bulk density values is also attributed to the enlargement of the pellets, resulting from the increase in pellet volume with the increase in moisture content (McMullen et al. 2005). This shows that many different parameters need to be adjusted for pellet production, so as to obtain acceptable bulk density values. A higher bulk density is generally desirable, as the more compact the material is, the less costly it becomes in terms of storage and transportation. It is worth mentioning that bulk density is usually associated with energy density, an important parameter that also assesses the pellet quality. It is defined as the energy content of the biomass material per unit volume, often expressed in GJ/m^3. Different types of biomass possess different levels of energy density. For example, miscanthus energy density is found to bet 2.05 GJ/m^3 (Whittaker and Shield 2017), whereas beech and spruce energy densities are 4.5 and 3.03 GJ/m^3 respectively (Arshadi et al.,2013; Finell et al. 2009; Holm et al. 2006, 2007). However, these values are highly increased upon densification, where pellets generally are found to have energy densities that vary from 9.8 to 14 GJ/m^3, which shows the importance of the densification process (Pellet Fuel Institute 2010). This variation is related to the original bulk density of the biomass used, in addition to its lower heating value, since energy densities are affected by these two parameters.

5.3. Effect of particle size on durability

Particle size is a significant factor that also affects pellet durability. Nevertheless, the effect of particle size on durability differs, depending on the type of biomass raw material used. In the study by Castellano et al. (2015), different samples were studied, including pinus pinaster (PIN), chips from Eucalyptus camaldulensis (EUC), pyrenean oak logging residues (OAK), chips from short rotation coppice (SRP), logs from a commercial crop (POP), vine shoots (VIN), aerial parts of oats (OAT), triticale (TRI) and rice straw (RIC). It was noted that some hardwood species were not affected in terms of durability when changing the milling size, unlike the poplar samples that achieved higher mechanical durability when minced at 2 mm. Pine and herbaceous materials milled at 2 mm also resulted in stronger pellets, as shown in Figure 3.

The data presented in Figure 3 agree with previous studies, such as that of Kaliyan and Morey (2008), which state that using finely ground biomass produce more durable pellets, compared to using coarsely ground biomass. Comparable outcomes were recorded by Esteban et al. (2006) and Shaw et al. (2008), where pine logging residues with 4 mm and 3 mm milling screen sizes were used to obtain advanced mechanical durability levels, compared to milling at 8 mm. Similarly, Shaw et al. (2008) described obtaining wheat straw and poplar wood pellets with advanced mechanical resistance when using finer particle sizes.

On the other hand, Serrano et al. (2011) did not find any noticeable differences in the mechanical durability of barley straw pellets when the raw materials were milled at 4 and 7 mm, even though Castellano et al. (2015) reported that a 2 mm milling screen size had a helpful effect on the mechanical durability with respect to a 4 mm milling screen size. Tabil and Sokhansanj (1996) also studied alfalfa pellet durability, and found that using different hammer mill screen

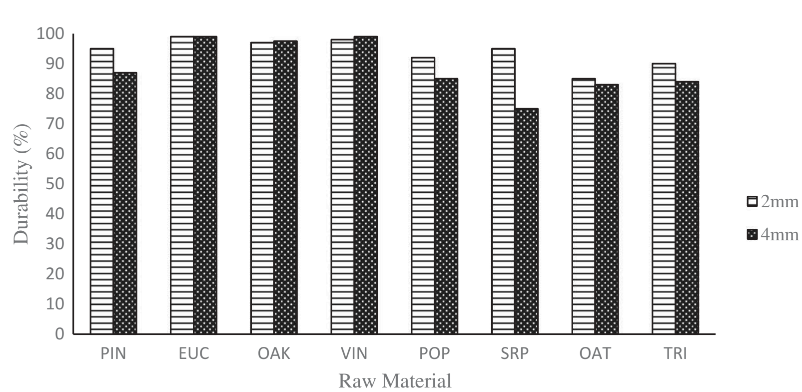

Figure 3. Durability of different raw materials at two particle sizes (note: Castellano et al. 2015).

sizes (anything between 3.2 mm and 6.5 mm) does not affect pellet durability.

In general, the lesser the particle size, the better the pellet durability reached (MacBain 1966). According to Theerarattananoon, Xu, and Wilson (2011) and Franke and Rey (2006), particle sizes ranging from 0.5 to 0.7 mm is recommended for good quality pellets. Payne (1978); Payne (2006) reported different levels of particle size that might reach a properly good pellet quality, noting that the effect of the inter-particle gaps should be minimalized. Fine particles typically absorb additional moisture related to larger particles and, hence, are capable of undergoing an advanced degree of conditioning. Moreover, the presence of large particle sizes is usually considered disadvantageous, due to their high tendency to crack and break down, which will eventually cause failure. Although more durable pellets are obtained by finer particles, fine grinding is unfavorable due to its higher cost.

Improved pellet qualities may be obtained using a variety of particle sizes, since they allow for inter-particle connection with almost the absence of inter-particle gaps. As for the differences in the results of the studies reported for pellet durability, based on the effect of decreasing or increasing the particle size, this can be explained by the use of industrial pellet mills in some studies such as Puig-Arnavat et al. (2016) and Theerarattananoon, Xu, and Wilson (2011), while using laboratory scale single pellet press units by others such as the study of Niedziolka et al. (2015) and Puig-Arnavat et al. (2016). Figure 4 presents a recommended proposal of feed particle size distributions, according to Payne et al., 2006.

This distribution was also adopted by Stelte et al. (2012), which in turn suggested that the wide difference in the particle size is recommended for obtaining higher pellet quality (Payne 1978; 2006). However, the presence of large quantities of fines (defined as particles with diameter smaller than 0.5 mm) may sometimes have an undesirable effect on pellet quality such as in the study of Maraver et al. (2015).

6. Types of binding mechanisms

In order to understand the mechanism through which the bonding of particles occurs during the pellet formation process, it is necessary to study the system under both microscopic and macroscopic conditions. According to Chung (1991), two different criteria for strong adhesion can be derived at the microscopic level: (1) an intimate molecular form of contact and (2) an extensive attractive strength associated with least potential energy. Once the maximum attractive strength and the minimum potential energy are close to each other, chemical bonds are made. Increasing the molecular interaction between any two groups of molecules may enhance the adhesion process, according to Chung (1991). This may be achieved by heating the system to temperatures higher than the glass transition temperatures in the presence of solvents (i.e. water). When amorphous materials pass through a transition from a solid glassy to a soft elastic state, glass transition is hence mitigated (Chung 1991), so that the material properties such as viscosity and modulus of elasticity could change dramatically (Chung 1991). Biomass cell contents may be activated by glass transition in biomass particles during the pelleting process. This consequently results in increasing pellet durability. Hence, glass transition conditions can increase the mobility of natural binding components, and assist the plastic deformation of particles.

On the other hand, there are two mechanisms through which binding forces between particles form at the macroscopic level (Pietsch 2001; Rumpf 1962), represented by bonding with and without a solid bridge among particles. There are many methods through which solid bridge type binding mechanisms may take effect (Pietsch 2001; Rumpf 1962). The process of cooling/drying densified products is usually the main mechanism through which solid bridges are formed (Kaliyan and Morey, 2008). Solid bridges may also occur from the molecular dispersion that may happen between the particles, specifically at the points of contact, associated with high pressures and temperatures. In addition, solid bridges can grow due to the crystallization of elements, chemical reactions, hardening of binders, and the change of the state of liquid components to solids. (Kaliyan and Morey, 2008)

In the absence of a solid bridge, particles may bond with the help of attractive forces between solid particles. Solid particles stick to each other through short-range forces (Kaliyan and Morey, 2008) such as molecular valance forces (i.e., free chemical bonds), hydrogen bridges, van der Waals' forces, electrostatic and magnetic forces. Molecular valance forces only take effect when the inter-molecular distance is around 10 Å. Unlike valance forces, van der Waals' forces are considered to be the most effective forces that contribute to all attractive and adhesive effects of molecules at a distance less than 0.1 μm. As a result of milling or friction within the particles, excess charges may be generated, so that electrostatic forces become very helpful in the binding process. The

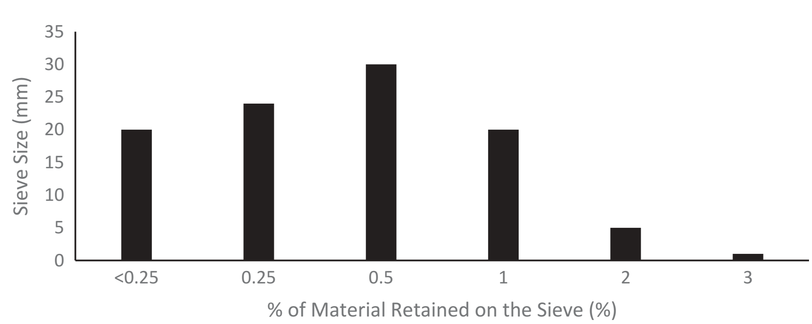

Figure 4. Typical particle distribution (adapted from Payne 2006).

presence of magnetic forces may also contribute to further particle bonding. When particle size, or inter-particle distance increases, the efficiency of short-range forces rapidly decreases (Pietsch 2001; Rumpf 1962).

Talking about corn stover and switchgrass, the glass transition occurs at temperatures ranging from 50°C to 113°C (Kaliyan and Morey, 2009a), whereas the average glass transition temperature of them is 75°C, for a moisture content ranging between 10% and 20% (Kaliyan and Morey, 2009a). Meanwhile, the plastic deformation of lignin is activated at temperatures of 60°C to 90°C, within the average glass transition temperature of corn stover and switchgrass, associated with low compaction pressures (Back 1987). On the other hand, it is very important to plasticize the wood polymers at temperatures above their glass transition temperatures, in order to have a sufficient bonding area for wood pellet formation. Hydrogen bonding at the lignin and cellulose surface areas is believed to be a major form of bonding, during the process of press-drying of wood, in addition to covalent bonds between the wood polymer chains during compaction at high temperatures (Back 1987).

The adhesion of biomass particles is highly affected by the contents of protein, pectin, stem material as well as the moisture content (Rehkugler et al., 1969). Durable densification from agricultural biomass materials, for instance, hay, can be achieved through the squashing, adhesion the interlocking of stem and leafy materials. Specifically, the moisture content in Lucerne hay increases the degree of adhesion because of the increased maceration of stem and leafy materials, and the increased extraction of protein and pectin from plant cells (Rehkugler et al., 1969).

Kaliyan and Morey (2008; 2009a) explained the effect of the presence of different levels of natural binding components on the durability of densified biomass products. In order to make use of their binding efficiency, natural binders such as fat, starch, lignin, and protein can be activated through high moisture contents, elevated temperatures or even steam (Kaliyan and Morey, 2009b; Back 1987; Rehkugler et al., 1969; Nyanzi and Maga 1992; Slade, 1993; Thomas et al., 1996). Figure 5 demonstrates natural binding process within corn stover. After introducing the pressure through the densification process, the natural binders were pressed out of the biomass cells. Further size reduction of biomass particles also caused a very light expression of protein compounds on the particles surface, which in turn is also acting as a natural binding agent.

Moore (1965) reported that water, which is usually used as a binder and a lubricant is especially helpful in densifying mixtures that have water-soluble components such as sugars, starches, sodium phosphate, and ash. A thin layer of water around the particles can strengthen and enhance van der Waals' forces. Moreover, water induces the formation of bonds via capillary absorption between particles (Thomas et al., 1996). In addition, water can result in many chemical and physical changes when coupled with heat, such as the denaturation of proteins, and the consecutive recrystallization of sugars and salts, which in turn affect the binding properties of biomass particle (Thomas et al., 1996). At moistures levels >20% w.b., the cell structure stays intact and pelletizing does not occur (Pietsch 2001).

At the macroscopic level, interparticle bonding could take place through a solid bridge or through inter-molecular attractive forces in the absence of solid bridge (Pietsch 2001; Rumpf 1962). Corn stover and switchgrass molecules may experience pressures of up to 100–200 MPa in roll-press pelleting machines (Kaliyan and Morey, 2009b; Dec 2002; Dec, Zavaliangos, and Cunningham 2003). After performing micro-structural analyses, it was proven that applying high pressure of 100–200 MPa to corn stover and switchgrass molecules in the pelletizing process can bring them close together resulting in enhancing the inter-particle attractive forces, and the formation of solid bridges as a result of pressuring the natural binders within the pellets. Cooling after pelletizing under pressure will harden the solid bridges and hence the pellets will acquire strength and durability.

7. Types of binders used

It is very important to cut down the energy requirements of pelletization processes, as well as reduce any resulting greenhouse gases emissions. Therefore, pellet binders are often used to improve pellet thermal conversion properties. Moreover, binders are also used to enhance pellet durability, and decrease any wearing effects on the pellet die. According to current European specifications (European Pellet Council 2013), the total amount of binders used in any pelletization process can reach a maximum of 2% of the pellet's total mass, which also applies for pellets derived from biomass raw material such as starch, flour or vegetable oils.

Wood pellets may either rely on organic or inorganic binders; nonetheless, the type of binder material is often

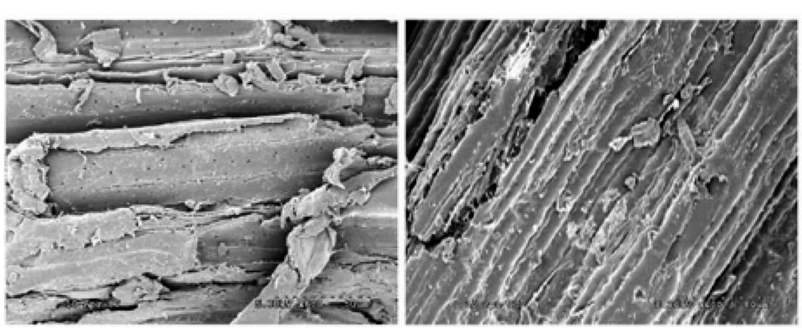

Figure 5. Natural binding within corn stover (adapted from: Kaliyan and Morey, 2009b).

chosen based on economic and environmental aspects, while ensuring enhanced pellet performance. For instance, when corn starch and molasses are used as binders, a higher durability for wood pellets is observed (Norway spruce and Scots pine) (Erlich et al. 2005; Yin et al., 2012). However, from an environmental perspective, these binders are not considered a good option, due to the indirect GHG emissions. The animal feed pellet industry often uses sugary and starchy binders for wood pellet production. In general, starchy binders (eg. corn or potato flour), as well as water-soluble carbohydrates, improve biomass pellet durability. Moreover, the durability of wood pellets increases by 10–20% when sugary binders (eg. sugar, molasses, cassava) are added.

Proteins that plasticize under heat effects generally enhance pellet durability, and are considered effective binding agents. Ahn et al. (2014) studied rapeseed flour, coffee meal, and lignin compounds that are high in protein and starch content. It was observed that the durability of larch and tulip tree wood pellets was enhanced upon adding such proteins. Since tulip pellets have a lower lignin content, and are inherently less durable that larch pellets, adding a value of binder between 2% and 10% continuously improved tulip tree pellets, while had no real effect on larch pellets Ahn et al. (2014). Another study considered the effect of binding agents on a blended form of miscanthus and sawdust pellets (Whittaker and Shield 2017). Since miscanthus is reported to be low in lignin, a high durability (equivalent to 97.5%) was only accomplished after adding 2% of potato starch binders (Whittaker and Shield 2017). However, when examining a 2.2% and 5.8% blend of waste vegetable oil with wood pellets, the result was increased energy value of the produced pellets but decreased durability (Mišljenović et al. 2015). Hence, it was found suitable to use oil-based coating agents after pellet formation, so as to enhance protection from external moisture, and increase the lower heating value.

Lignin, a non-food based additive, can be used for pellet feedstocks where the additive's relative content is low. For instance, a 0.8% and 1.4% increase of pellet durability was observed in Scots pine and Norway Spruce, respectively, after a 2% addition of Kraft lignin and lignosulphate (Kuokkanen et al. 2011). Even though glycerol is not effective when it comes to enhancing pellet durability, it does show lower ash content and increased higher heating value when used for wheat, barley, oat, and canola straw pelletization. Algae, on the other hand, result in improved compressive strength of Miscanthus pellets. However, adding more than 20% causes a decrease in the lower heating value. Recently, inorganic silicate-based binders are becoming more prevalent, due to the numerous advantages it offers, including ease of handling, better cost, safety, and reduced emissions of NOX and VOC's.

Sodium silicate is a special binder that can either be used as a matrix, film or a chemical binder. In order to reach a decent pellet strength, agglomerated materials often use binders such as sodium silicate, which fit in those three different categories. For instance, sodium silicate can be used as matrix binder in the cement industry (Mike McDonald and Hamilton 2009). Sodium silicates which have 45–65% water by weight facilitate the formation of a strong, rigid, glassy film, which assists in enhancing pellet strength. The viscosity, concentration, temperature, film thickness, and relative humidity are all factors that contribute to the rate of drying of sodium silicates during pelletization. It is possible to enhance the penetration and the adhesive forces of soluble silicate through the use of surfactants/wetting agents. Such agents also help strengthen the bond between pellets, by moderately increasing their degree of dispersion over the pellet surface. Sodium silicate dispersion is usually larger when the molecules can penetrate through smaller openings, with the aid of reduced surface tension.

Both silicate and lignin are considered the most popular binders in the pellet industry, can sequester metals, may be used dispersants, as well as in many other applications. Nevertheless, a recent study has reported some complementary applications for many lignin and silicate compounds, especially for calcium lignosulfonate (McDonald and Hamilton 2009). Calcium lignosulfonate may be found in both solid and liquid form (~50% solids). However, both forms of calcium lignosulfonate can be used with silicates. Even though potassium or lithium silicate may sometimes be used for particular applications, sodium silicate is the most used binder compared to other alkali silicates, due to its competitive price. When sodium silicate is considered undesirable for a certain application, potassium silicate is utilized. For example, potassium silicate was found suitable for welding rod applications and in acid-resistant cements (McDonald and Hamilton 2009). In addition, potassium silicate is currently under study as a potential binder in the fertilizer industry; increased solubility of potassium silicate can generally improve pellet strength.

As for lithium silicate, its properties lie in between those for sodium silicate, and organic ammonium silicate. The difference between lithium and sodium silicate is in being able to stabilize more silica in the colloidal state, due to the fact that a hydrated lithium ion is bigger than a hydrated sodium ion. Even though lithium silicate is considered expensive, it can result in better adhesion than that of sodium and potassium silicates. Hence, a minor amount of lithium silicate mixed with sodium or potassium silicate can enhance adhesion considerably (FFalcone 1982). In addition, unlike sodium silicate, lithium silicates are companionable with water-miscible organic solvents, and are slightly companionable with non-polar solvents, which results in better bonding when used with materials that are covered with organic solvents.

Some other commonly used binders are lignosulphonate, starch, dolomite, corn or potato flour, and some vegetable oils (Tarasov et al., 2010). Lignosulphonates is a water-soluble anionic polyelectrolyte polymer, and is mostly used as a binding agent in wood pellets manufacturing. Starch is usually made from two different polymers, mainly amylose and amylopectin. This binder can be found in many different shapes and granular sizes, which greatly affects its distribution on wood biomass material, often resulting in pellet abrasion. Other binders, such as vegetable oil, are used to provide enhanced lubrication conditions through the pelletization procedure.

Bentonite (an absorbent made from aluminum phyllosilicate clay) is lately being used as binders for iron ore pellets, since this clay material is capable of absorbing water through its structural layers, leading to gel formation (Zambrano et al. 2013). Moreover, lime is a very popular binder in pelletization when it comes to enhancing the quality of green pellets. In addition to the lime and clays, various compounds are being considered as potential binders for pelletization such as: hydrated magnesium chloride, magnesium oxide, and calcium chloride. Organic reagents are often favored, since they minimize residue formation of the pelletization process. This is particularly essential when manufacturing certain types of steel pellets, where the amount of impurities should be kept minimal. Moreover, even though inorganic binders are more economic than organic types, the equivalent amount organic binder needed per unit mass of raw material is much lower, compared to inorganic binder requirements. Moreover, organic products are silica-free, leaves no residues, and their addition does not affect the iron concentration. However, it should be noted that these binders are often costly, highly hygroscopic, easily polymerizing in water and produce pellets with lower compressive strength, both dry and green.

7.1. Effect of binders on wood pellet properties

Moisture content, particle density, number of fines, mechanical strength, calorific value, and ash melting point are all major physical and thermal parameters that affect wood pellet quality. Each of those properties may be adjusted by means of binders with different concentrations throughout the pellet manufacturing process.

For instance, calorific value of the pellets is a major factor that affects the quality of the pellets produced, since it gives an indication of the amount of heat that will be produced from the pellets when subject to thermal conversion. However, it was noted that the calorific values of the biomass raw material are found to be lower than that of the produced pellets. For example, the calorific value of miscanthus as a raw material was found to be 12.1 MJ/kg (Whittaker and Shield 2017). However, this value was increased to 18.5 MJ/kg upon pelletization (Whittaker and Shield 2017). Also, wheat, barley, rape, and maize straw raw materials had calorific values of 17.1, 16, 14, and 16.3 MJ/kg, respectively, whereas the same materials in a pelletized from had calorific values of 18.83, 18.88, 18.1, and 18.8 MJ/kg, respectively (Mani, 2004; Mani,2003; Serrano et al. 2011; Wongsiriamnuay PK, 2002) . Other biomass types were also showing the same behavior upon pelletization, where the calorific value of oats increased from 17.2 to 18.9 MJ/kg, pine wood's calorific value increased from 16 to 18.8 MJ/kg, and larch's calorific value increased from 17.6 to 19.2 MJ/kg (Adapa et al., 2009; Arshadi et al., 2008; R. Samuelsson et al. 2009; Arrieche et al. 2011) .

In addition, since moisture content is an important pellet property, water is usually added to the biomass raw material at the beginning of the manufacturing process. The optimal moisture content is affected by the nature and quantity of the binding agent used. When dolomite is used as a binding agent, the ability to absorb water increases significantly, while wheat starch does not affect water consumption for pellet production (Tarasov et al., 2010). A moisture content between 12.5% and 13.0% is needed when starch is used as a binding agent, while a moisture content ranging between 9.0% and 10.5% is needed when lignosulphonate is used. Moreover, mixing an amount of 1% by weight wheat starch with 1% by weight oxidized corn starch along with wood biomass raw material that contains 12.1% moisture content, the wood pellet moisture content decreases to 7.6%. In this case, the more the dosage of the binding additive used, the lesser the moisture content value obtained in the final pellet product. In contrast, when lignosulphonate and maize starch are added simultaneously (1% lignosulphonate and 1–4% maize starch), the final wood pellet moisture content declines by 0.5%. Therefore, it can be concluded that the addition of starch has a stronger effect in decreasing the final wood pellet moisture content, with respect to lignosulphonate. However, if the amount of starch exceeds an amount of 4% by weight, the final pellet product obtained will be extremely dry (Chhiti and Kemiha 2013).

Moreover, the power consumption of wood pellet production process is greatly affected by the binding agent utilized, particularly maize starch and lignosulphonate, which provide desirable power consumption when compared to other binder options. The specific energy intake needed for the manufacturing of poplar wood pellet depends strongly on the binding agents added (Tarasov et al., 2010). As a base case (i.e., without adding binders), the specific energy consumption for producing poplar wood pellet in the study of Tarasov et al. (2010) was estimated to be 138 kWh/ton dry biomass.

When it comes to durability, lignosulphonate results in higher mechanical durability when used for pelletizing, compared to maize starch. On the other hand, using corn starch and dolomite as binders result in a decrease in the wood pellet particle density.

In the work of Tarasov et al. (2010), it was stated that in the absence of a cutting blade, starch binding agents increase the wood pellet bulk density. Moreover, when raw materials of smaller particle size and lower moisture content are used, the bulk density of the pellet increases (Tarasov et al., 2010). For instance, binders such as lignosulphonate (in addition to different types of starch) reduce the moisture content of the wood pellets and consequently increase their bulk density (Tarasov et al., 2010). Employing high process temperatures and pressures increase the pellet bulk density. According to Tabil (1996) and Samuelsson (2009), a higher moisture content results in a lower pellet bulk density.

Moreover, it has been experimentally proven that the amount of metal present in the binding material plays a key role in the attained bulk density of pellets made of softwood. For example, an average increase of 22 kg/m^3 in the bulk density is observed upon the addition of 5% bark binder, which contains concentrations of metals 10 times more than stem wood (such as aluminum, iron, and sodium) (Ishii and Furuichi 2014; Samuelsson et al. 2009).

A study conducted by Ahn et al. (2014) used Larch (LAR) and Tulip tree (TUT) as raw materials for pellet production. The durability values that were reported in this study for pellets produced with only LAR and TUT sawdust were 96.9% and 95.1%, respectively. Binders such as rapeseed flour (RF), coffee meal (CM), bark (BK), pine cone (PC), and lignin powder (LP) were then added, to enhance the durability of LAR and TUT pellets (Ahn et al. 2014). In case pellet strength and durability are not in agreement with quality standards, binders of amounts that vary between 0.5% and 5% (by weight) may be used to enhance pellet quality, and reduce any durability variations.

For LAR pellets, it can be observed that rapeseed flour (RF), coffee meal (CM) and lignin powder (LP) binders resulted in the highest durability values, while the lowest durability values were observed when bark (BK) was used as a binder. Similarly, the highest durability values were observed for TUT pellets when produced with RF and LP, then CM, while the least durability values were obtained using BK and PC as binders. The high durability attained for LAR and TUT pellets that are manufactured using RF and LP can be attributed to the auto binding properties of those two binders. Unlike LAR/CM pellets, the durability of TUT/CM pellets was reported to be considerably lower than TUT/RF and TUT/LP pellets, even though CM contains high quantities of lignin (41.7%) and protein (11.6%). Also, the relatively high fat and oil content of CM binders (22.7%) compared RF binders, (11.7%) results in a decrease in pellet durability, since oil by itself lubricates the walls of the die where particles are fed, minimizing by that the die pressure (Briggs JL 1999). Moreover, a considerable decrease in the durability of LAR pellets is observed when BK or PC are utilized as binders in the pelletization process, and there exist very few positive effects on the TUT pellets durability when those binders are used. This is due to the fact that BK and PC binders are associated with a bigger particle size, and a higher moisture content compared to LP. Moreover, the tannin compounds contained in BK and PC are not activated at low temperatures. Therefore, high temperatures are always desired to be able to utilize BK and PC as binders in wood composite materials.

On the other hand, the durability values attained for TUT pellets were directly proportional to the overall quantity of the binder; an improved durability was observed for TUT pellets, when the amount of the binder used is increased. This observed effect is a result of the intermolecular and magnetic forces inside the TUT pellet itself that enhance the adhesion mechanisms within the pellet.

Regarding the effect of the amount and type of binder used for LAR pellets, it was observed that adding LP and CM binders to LAR pellets with a quantity ranging from 2% to 10% gradually improved the durability values of LAR/LP and LAR/CM pellets, starting from an initial value of 96.9% in the case of the absence of binders. In contrast, blending LAR particles with RF binder quantities that are between 2% and 10% produced LAR/RF pellets with irregular durability values. Then again, adding PC and BK binders to LAR pellets with a quantity ranging from 2% to 10% steadily decreased the durability values of LAR/PC and LAR/BK pellets (Ahn et al. 2014).

Even though this trend was observed by Ahn et al. (2014), Bradfield (1984) and Yaman et al., (2001) state that adding BK and PC could generally improve the quality of densified products. Such contradictory results from different literature sources, may, in fact, be due to the small chemical reactivity of tannin present in BK and PC binding materials, as well as to their higher moisture content and larger particle size when compared to LP binders.

As for TUT pellets, the higher the binder quantity utilized, the longer the durability observed. In particular, increasing quantity of LP binders resulted in higher durability values of TUT pellets.

In case of dealing with TUT/BK and TUT/PC pellets, increasing the quantity of BK and PC slightly improved the pellet durability, compared to LAR/BK and LAR/PC pellets. This is believed to be a result of differences in specific gravity values for LAR and TUT. Therefore, the durability of TUT/BK and TUT/PC may be slightly enhanced when adding the amount of BK or PC from 2% to 10%.

Meanwhile, LP revealed the best results for improving the durability values of LAR and TUT pellets. The adding of RF, CM, and LP binders to LAR or TUT samples formed pellets with acceptable durability values in some case, due to the presence of high protein and lignin contents. To study the influence of both the type of binder used and the feed moisture content (MC) on the durability of LAR and TUT pellets, feed moisture content was adjusted to 13–19%. In particular, LAR and TUT pellets with a feed moisture content anywhere between 17% and 19%, were reported to have the highest durability values (Ahn et al. 2014).

On the other hand, in the study of Lu et al. (2014), lignosulfonate and bentonite (each of 2% by weight) were independently added on wheat straw milled particles. Crude glycerol was added to the mixtures as a binder, so that enhanced properties were observed, due to the increased tensile strength of the formed pellets. Nonetheless, the addition of crude glycerol to the mixture of the pellets caused an increase in the density of the pellets obtained, associated with higher heating values, compared to pellets that are manufactured in the absence of binders, and pellets that are blended with other binders. However, adding bentonite and lignosulfonate to the pellet's mixture lowers the heating value and the specific energy consumption of the pelletizing process, and increases the pellet density and tensile strength. For instance, in the case of dealing with wheat straw pellets without addition of any binder, it was noticed that the tensile strength was 0.84 MPa. However, this value increases to1.63 MPa when adding binders with an amount of 20% (by weight). It is recommended that compressive forces be given more attention when assessing pellet quality. As a matter of fact, when pellets are packed and prepared for transportation and distribution, they are subject to compression rather than tensile forces, due to continuous particle collisions. Therefore, compressive forces can be more effective for assessing pellet performance than the tensile ones. Moreover, the pellet heating value is a key factor that stimulates the binding mechanisms within pellet particles. Kaliyan and Morey (2010) found that increasing the heating temperature used for corn stover pelletization from 25°C to 75°C increased the durability from 75% to 97%, as shown in the SEM images provided in Figure 6.

Finally, it is worth mentioning that recently, catalysts are also being used to enhance the properties of the produced pellets. For example, in the study of Sun (2017), the thermal

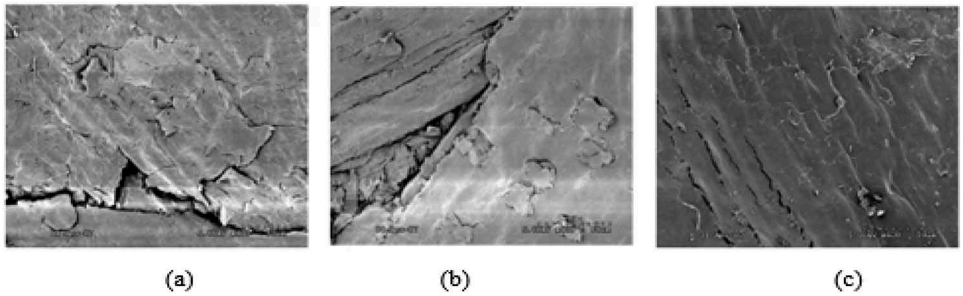

Figure 6. SEM image of corn stover pellets (a): T = 25°C, and Du = 75%. (b) T = 25°C, and Du = 96%. (c) T = 75°C, and Du = 97%. (adapted from: Kaliyan and Morey 2010).

properties of pine wood were investigated upon the addition of CaO/ZSM-5 catalyst, where results showed that the pyrolysis yields of the pine wood with catalyst where higher than pyrolyzing the pine wood alone (Sun, 2017). In addition, the work of Zhang (2014) showed that the use of Fe(III)/CaO catalyst increased the vapor yields obtained upon the pyrolysis of biomass, caused by the decrease of the amounts of oxygenated compounds. Veses et al. (2014) investigated the pyrolysis of wood using CaO and CaO· MgO catalysts at 450°C. Their findings showed that the catalysts used upgraded the quality of the bio-oil obtained from the pyrolysis process. It was also found that nickel oxides can be used as catalysts. However, the work of Abu Tarboush et al. (2012; 2015) proved that oxidizing asphaltenes onto nickel oxides nanoparticles did not possess catalytic activity that pertains to the oxidation of adsorbed particles, and only improved the contact of the adsorbed asphlatenes with its nearby surroundings.

Out of all what has been previously mentioned, it can be concluded that many factors affect the pellet durability parameter. However, it should be noted that this parameter only considers the ability of the pellet to resist shocks, which is determined according to the pellet conditions (such as moisture content). Obtaining pellet durability values are much more useful than the characterization of the pellet in terms of tensile and compressive strengths. It was found extremely important to introduce a new lumped parameter that assesses the quality of the pellets, by taking into consideration all the following useful parameters that affect the pellet quality: the mechanical strength (MS), the heating value (HV), the feed composition (FC), the durability (Du) and the yield quantities upon thermal conversion (Y). By defining this parameter as the quality of the pellets (Q), it can be easily addressed as follows:

$$Q = xMS + yHV + zFC + wDu + mY \quad (1)$$

Equation (1) can then be used to determine the pellet quality parameter (Q). However, it should be noted that the constants (x, y, z, w, m) may be obtained using multiple linear regression methods. For this, sufficient data must be obtained regarding how pellet behavior is affected by the various factors listed above. Moreover, it should also be noted that some factors must be given more importance than others, when quantifying pellet qualities. For instance, the parameters associated with the mechanical strength and heating value (x and y) should be assigned the highest weighing factors, since mechanical strength associated with compressive forces affect the mechanical properties of the pellet, while the yields affect the heating behavior of the pellets when burnt, which consequently defines the main target of palletization, in addition to playing a major role in identifying the corresponding heating value of the pellets. It should be also noted that the feed composition takes into consideration the moisture content of the feedstock in addition to the particle size distribution, and whether binders and catalyst are used or not.

8. The pelletization process

The pelletization process mainly consists of three major steps: raw material pre-treatment, pelletization, and post-treatment. Hence, following raw material preparation stage, the pelletization process then follows as shown in Figure 7.

During the raw material pretreatment stage, special care should be given to feedstock storage, since it is essential to maintain the biomass raw material at an acceptable level of purity, by keeping it in a dry environment using an adequate feedstock storage system. Moreover, the removal of the undesirable impurities is very essential. The raw material should first be filtered, then grinded. In order to get rid of any metal fragments or stones, it must then undergo a magnetic separation process to remove the metal remnants and other impurities from the biomass bulk. It is very important to get rid of such impurities, to avoid any serious damage to the mechanical equipment. Following impurity removal, a size reduction process is implemented, in which the raw material is minced to a uniform size of 6 mm diameter, in order to prevent the blocking of the holes of the pellet mill. In addition, having small raw material feed particles with a large surface area during pelletization would result in a higher density, and stronger pellets. Moreover, even the moisture optimization process during the drying phase becomes easier when having a narrow raw material particle size distribution.

Following size reduction, the raw material may be then transported via screw conveyors. Another alternative is using a fan followed by a cyclone separator that separates air from the milled material. However, the first technique is favored due to its lower cost. In order to obtain high-quality pellets, the raw material should maintain a moisture content between 10% and 20%. Drying solid biofuels is necessary, since wet raw materials result in low energy efficiency and high emissions of hydrocarbons and

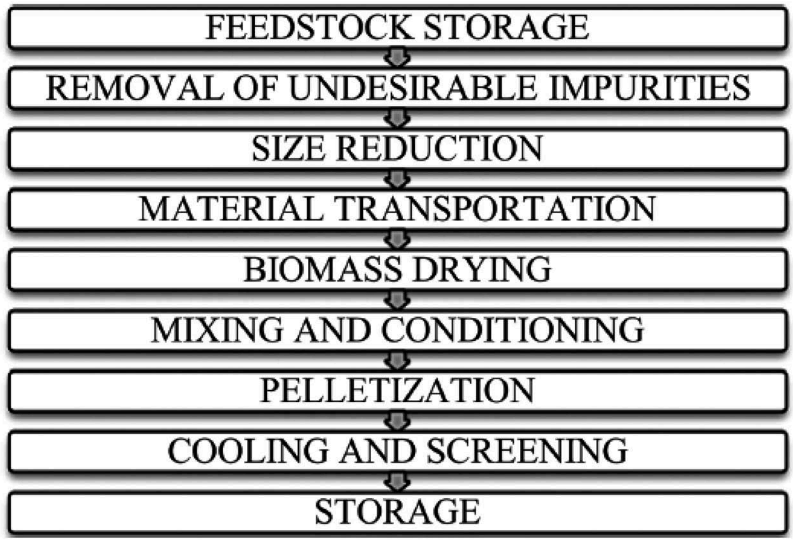

Figure 7. Stages of the pelletization process (adapted from WIT transactions on state of the art in science and engineering, Vol 85).

particles. Besides, it is essential to use an adequate dryer. Since drying equipment usually corresponds to the largest capital expenditure in a pellet production plant, this must be appropriately accounted for in the pellet production costs.

Not all raw materials require mixing. However, mixers can be used after size reduction and drying, if needed, in order to obtain a consistent and homogeneous material blend. Moreover, adding suitable binder materials is extremely important, to enhance the properties of the final product. As mentioned previously, binding agents can promote durability, enhance some of the pellet physical qualities, increase pelleting efficiency and decrease energy costs. The binder agents are chosen based on the raw material used in the pelletization process, to ensure that the most desirable properties of the final pelletized product may be attained.

Pelletization is conducted in pellet mills, identified as pellet presses or extruders. Pellet mills work by generating a high pressure, so as to direct the raw material through the openings of the die. As the pressure increases, the friction increases, and the temperature of the raw material also rises. This results in a decreased moisture content, softening of the lignin and reshaping of the fiber into pellet form.

Pellet mills are classified either as flat or round pellet mill, according to the shape of the die. Flat die pellet mills contain a die with holes, where the raw material is placed on the top of the flat die. Once the die starts rotating, the raw material is pressed and passes through the openings of the die, and the pellets are finally cut. Such pellet mills are utilized for small to medium scale pellet productions. However, round die pellet mills, on the other hand, have circular openings along the die placed vertically. The raw material is put in the middle of the die, and is then evenly distributed upon operation. After that, the rollers squeeze the material through the openings, and the obtained pellets are cut by blades located outside the die.

At the end of the pelletization process, it is essential to cool the produced pellets, before leaving the production plant for the sake of pellet strength and durability. Pellets are usually air cooled once they are removed from the extruder, in order to promote the solidification of the lignin which strengthens the pellets. Once the pellets have cooled off, they are screened to get rid of any fine material and dust. Any fine material is recycled back into the pelletization process. Screening is the last step in pelletization, after which the pellets are set for packaging.

9. Parameters affecting the pelletization process

Both pellet mills and roll presses are commonly used for densification processes, to produce durable pellets that are acceptable in quality, and good in shape. The following sections describe the variables that affect the densification process, since many different factors are involved.

9.1. *Effect of forming pressure*

High pressure conditions enable the activation of various binding mechanisms, mainly inter-particle bonding which occurs where natural binders such as starch, protein, lignin, and pectin in the feed are squeezed out of the particles. Therefore, high pressure conditions in the range of 100– 150 MPa for pellet mills, and between 100 and 200 MPa for roll presses, are favored in densification processes. Kaliyan and Morey (2008) studied the effect of pressure on the durability of grass hay pellets (mixed with 20% alfalfa), and it was noticed that adding pressure from 5 to 44 MPa, raised the durability from 5% to 91%.

Kaliyan and Morey (2008) also studied the forming pressure effects on the rice husk raw material. When the samples were first heated from 30°C to 225°C as a kind of pretreatment method, at a constant pressure of 68.6 MPa, the durability of alfalfa pellets raised from 7% to 72%. When increasing the pressure to 257.4 MPa, the durability increased from 40% to 99.8%. Moreover, tensile strength of alfalfa pellets was noticed to raise from 0.5 to 2.0 MPa when adding the pressure from 15 to 100 MPa. However, tensile strength dropped from 1.5 to 2.0 MPa, when adding the pressure over 100 MPa and up to 165 MPa . Therefore, higher pelletization pressures result in higher pellet tensile strength values, up to a point after which the values start to decrease.

9.2. Pellet-mill variables

In the pellet mill systems like the system in Figure 8, the dimensions of the die being used greatly affect the process. According to Heffner and Pfost (1973), a smaller die size results in a better gelatinization mechanisms in the poultry feed, and a better durability.

Hence, the greater the die length to diameter ratio, the better the durability values attained. However, a reasonable length to diameter ratio should be selected, since neither a small die nor an extremely long one is appropriate, otherwise, the pellet mill would be blocked. As for shear force magnitudes, those often depend on the friction constant between the feed and the die exterior. Franke and Rey (2006) proposed in their study that a long die can be effective for enhancing the durability of pellets that are made from raw materials with a high fat content. Also, the amount of material pelleted, the pellet bulk density, the pellet durability as well as the energy needed for compression, are all greatly affected by the die speed used (Tumuluru 2016). During the extrusion of feed pellets, the screw speed is observed to reduce viscosity, die pressure, and product temperature (Rolfe, Huff, and Hsief 2001).

Moreover, many studies have been conducted to investigate the effect of the distance between the roller and the die on the durability. In the studies of Apelt and Robohm, 1989, the mentioned effect was extensively investigated, and it was found that for a flat-die and a ring-die press, a greater pellet hardness and durability are observed upon increasing the gap-size to about 2–2.5 mm (Kaliyan and Morey, 2008). Nonetheless, a reduction in the pellet hardness and durability values were reported by the same study, upon increasing the gap-size to about 4–5 mm. This decrease may be attributed to sideway leaking of the feed mash, which mainly results in the destabilization of the feed crushing that occurs at the end of the roller and die. Durability data is obtained from the study by Robohm and Apelt (1989) and Bradfield et al. (1984), for pellets made from pig biomass material, in terms of the gap magnitude of the flat-die press, and summarized in Figure 9.

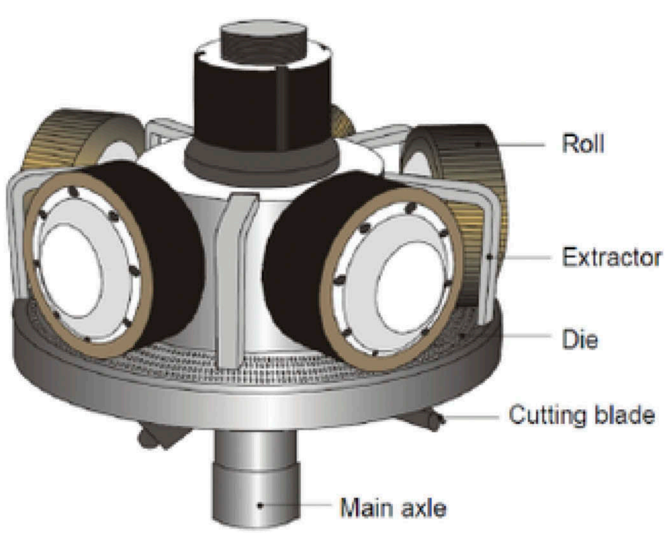

Figure 8. Pellet mill system (Stelte et al. 2011).

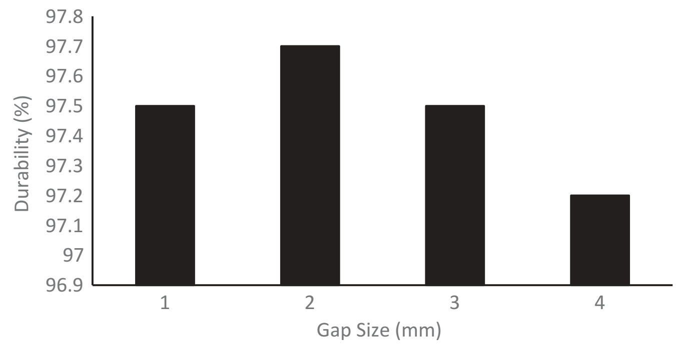

Figure 9. Variation of durability as a function of the gap size (generated from: Robohm and Apelt 1989).

Moreover, the durability of pellets (made from pig biomass) was reported to be 97.7% for a zero gap-size, and 97.9% for a gap-size of 5 mm.

It is also worth saying that increasing the electrical energy input, as well as the electric power used specifically by the motor of the pellet-mill, was reported to increase pellet durability, in many studies. For instance, Tabil et al. (2004; 2009) studied the effect of increasing the specific energy consumed by the pellet-mill motor, and found that the pellet durability values eventually increase. Moreover, Kaliyan and Morey (2008) proved that the durability of alfalfa pellets raised to 80% after when using 33.0 kWh/ton of biomass feed, after being 25% at energy input of 26.5 kWh/ton of biomass feed. In addition, according to Payne (1978), pellet durability is also affected by the amount of power input to the motor of the mill. It was concluded that a power of 10, 12–15, 20–25, 12, and 12 kWh/per ton of biomass feed should be maintained to produce high durability pellets (i.e., >92% Holmen pellet durability) specifically in the case of pellets from poultry, fin fish, and shrimp, respectively.

As for enhancing the hardness of the pellets, double pelleting is often utilized to improve the hardness and durability of pellets made from biomass raw materials with a high fiber or fat content. The process consists of two consecutive presses; one for compressing the raw material before entering the feed that is associated with a thin die, while pelletization process occurs in the second die, which is equipped with a thicker die (Tumuluru et al., 2016).

Regarding the expander, it is usually used for pre-densifying and mixing the feed mash, which would often result in a different structure of the feed particles. This process helps to enhance the pellet durability. By introducing shear stresses, the expander can increase the temperature of the raw material entering the feed to values above 100°C, hence enhancing the binding characteristics of the feed, and the durability of the formed pellets. In addition, increasing the pressure in the expander would result in a higher pellet durability. For instance, when the expander pressure increased from 20 to 30 bar, the durability of corn pellets increased from 85% to 89% respectively (Dec 1991).

Finally, the ripener is used during pelletization processes for feed conditioning. Before conditioning, the dwell time (time where pellets remain in the same stage) of the raw material in the ripener is around 15–30 min (Kaliyan and

Morey, 2008). A ripener is often utilized for the purpose of increasing the feed mash conditioning time, as well as for increasing the durability of pellets of molasses levels of 7–9% (high molasses), harshly milled particles and raw materials with a high moisture content.

9.3. Roll press variables

There are five main key factors governing the performance of a roll press: the screw feeder speed, the roller speed, the opening between the rollers, the pressure, and system's rigidity. Typical screw speeds in a roll press are in the range of 7.5–300 rpm, typical roll speeds are in the range of 1– 16 rpm (or 0.01–1.5 ms^{-1}), while a typical pressing force can range from10–150 kN. cm^{-1}. Table 2 summarizes the experimental work conducted by Komarek (1991) and Dec (2002) which investigated the effects of changing certain parameters on the strength and durability of the densified raw material.

According to Payne (2004), the roll pressing process efficiency mainly depends on the speed of the rollers and the screws and the applied pressure. Recent studies (Rambaliet al., 2011; Von Eggelkraut-Gottanka et al. 2002; Maier and Bakker, 1992) reported that roll press variables can greatly affect the properties of the produced densified material, especially in pharmaceutical applications. It was concluded by many of those studies that pressure is the most significant factor that contributes to the properties of lactose granules, then the roller speed, and finally the screws speed. Inghelbrecht and Remon (1998) conducted several experimental studies on four different types of lactose powders, under various hydraulic pressures conditions that are applied to the roll press, vertical screw feeder speed, and roll speed on the roller compacted granules. Table 3 summarizes the experimental results, which have been found to be optimal, as high durability granules were obtained at those reported conditions.

Moreover, according to Rambali et al. (2011), lesser pressure, smoother rollers, and a bigger gap dimension often produces stronger tablets. On the other hand, Von Eggelkraut-Gottanka (2002) stated that the smashing power of tablets made from herbal extract increased, after increasing the pressure introduced to the rollers. Those contradicting results, which have been reported by different studies, can be related to the variations in the mechanical characteristics of the raw material used.

Table 2. Effect of certain variables on the crushing (strength and durability generated from: Komarek 1991; Dec RT, 2003).

Parameter		Effect	
Pressure	120 MPa	Crushing Strength	225.6 N
	240 MPa		892.4 N
Roll force	50 kN cm^{-1}	Crushing Strength	75 N
	60 kN cm^{-1}		1300 N
Specific Energy to the Roll	15 kWh/t	Crushing Strength	313.8 N
	40 kWh/t		1333.7 N
Specific Energy to the Roll	5 kWh/t	Durability	74 %
	20 kWh/t		89 %

Post-production conditions have been also proven to significantly affect pellet quality. For example, the time of measurement, which contributes to a parameter called the green strength, is considered a key parameter in the post-production stage. Green strength is defined as the strength of pellets measured right after pellet production, while cured strength is defined as the strength of pellets measured within 1 week of pellet production (Kaliyan and Morey, 2009a). According to Payne (1987), dairy feed pellet hardness values were observed to be 78.5 N when measured immediately after cooling, increased to 131.4 N after 24 h. This hardness increase is a result of solid bridge formation during the lag time.

Cooling and drying are also important factors in the post-production stage. Following the pellet production process, the pellet temperature usually varies between 60 and 95 °C right after, and the moisture content is in the range of 12–18% (Koser, Schmalstieg, and Siemers 1982). To ensure safe storage, any excess heat and moisture should be removed. Therefore, pellets are usually air-cooled for about 4 to 15 min, to about room temperature and the moisture content is kept at around 0.5% of the initial moisture content of the raw material (Koser, Schmalstieg, and Siemers 1982). Pellets are usually stored under dry conditions, since increased moisture could significantly decrease the pellet compressive strength, and disintegrate some types of pellets made from oak sawdust, pine, and cottonwood according to Koser, Schmalstieg, and Siemers (1982). In addition to pellet strength, elevated moisture levels may also affect pellet durability. According to Fasina and Sokhansanj (1992), a 3–5% increase in moisture content due to storage under relatively high humidity conditions (70–90%) had detrimental effects on alfalfa pellets durability. However, in some cases, a 3–4% increase in moisture content could have a positive effect on the durability and strength, as it can strengthen the bond between individual pellet particles, as a result of the binding forces between water molecules (Fasina and Sokhansanj 1996).

10. The post- pelletization process (thermal conversion modes)

Following pellet production, and pellet characterization in terms of durability and bulk density, pellets may then be distributed and utilized in various ways. Pellets can be used thermally via three different pellet thermal conversion modes: gasification, pyrolysis, and combustion. Each of which is associated with different products, based on the quality of the pellets utilized. Both durable and environmentally friendly pellets with clean products are favored for thermal use. The following section presents an overview of pellet gasification, pyrolysis, and combustion, by highlighting the main contributing parameters of each process and the respective products. Based on the discussion provided, for the different pellet thermal conversion modes, the various environmental impacts associated with pelletization processes have been reviewed.

Table 3. Experimental Variables (generated from: Inghelbrecht and Remon 1998).

	Variable	Applied	Optimal
Quality of the Compacted Lactose Powders	Pressure	2.5–17.5 MPa	6.9 MPa
	Vertical screw feeder speed	100– 1000 rpm	1000 rpm
	Roll speed	3–13 rpm	7 rpm

11. Gasification

Gasification is a process through which organic carbonaceous resources are transformed into carbon monoxide (CO), hydrogen (H_2) and carbon dioxide (CO_2). Biomass gasification is widely used in energy industries, as it leads to the production of many beneficial yields such as syngas (synthesis gas), H_2, methane (CH_4) and other products. The gasification process itself does not involve combustion, it mainly involves reacting the organic resources under very high temperatures (600–1500°C) with a well-specified quantity of oxygen and/or steam (Chhiti and Kemiha 2013). The blend of the gases released through the gasification process is referred to as syngas. During a systematic gasification reaction, syngas should contain H_2, CO, CO_2, CH_4, H_2O, particles of higher hydrocarbons, impurities and probably some inert gases (Farzad, Mandegari, and Görgens 2016). The produced syngas has many applications especially in gas engines, to furtherly produce methanol (CH_3OH) and H_2, or improved to synthetic fuel by the Fischer–Tropsch process (Farzad, Mandegari, and Görgens 2016). Gasification reaction is mainly composed of different thermal processes: Drying, Pyrolysis, Combustion, and Reduction. Figure 10 summarizes an entire gasification process.

First off, drying is the process through which the moisture is removed from biomass material, before entering the pyrolysis stage. Pyrolysis involves subjecting the biomass raw material to high temperature conditions (usually above 240° C) in an environment void of air, and transforms it to char, tar gasses and liquids (Farzad, Mandegari, and Görgens 2016).

Combustion is the only net exothermic reaction in the gasification process, usually starts with the tar gasses or char from pyrolysis. The heat generated from the combustion step is needed for the upcoming processes (drying, pyrolysis, and reduction) and may be further collected by heat recovery systems. The reaction takes place according to equations (2) and (3):

$$C + O_2 \rightarrow CO_2 \; \Delta H = -393 {}^{MJ}/_{kg\ mole} \tag{2}$$

$$2H_2 + O_2 \rightarrow 2H_2O \; \Delta H = -242 \; {}^{MJ}/_{kg\ mole} \tag{3}$$

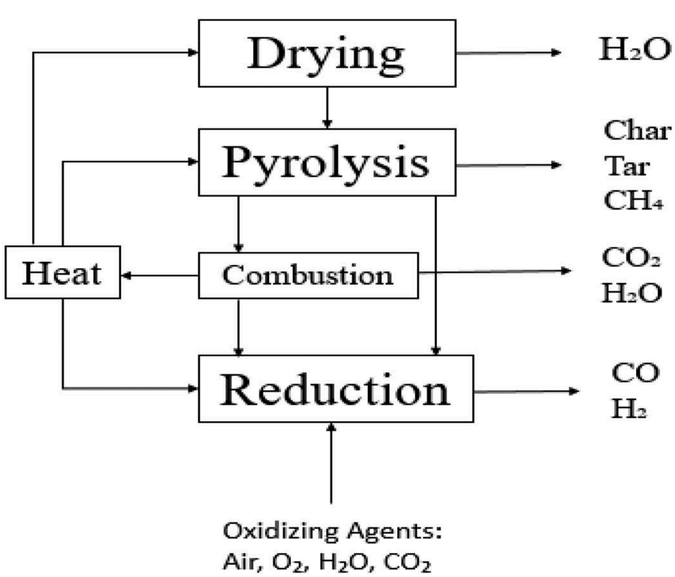

Figure 10. The processes of gasification.

Cracking in general also occurs, where decomposition of big complex particles to smaller ones under the effect of heat, like breaking down tar heavy molecules to smaller and lighter ones takes place, shown in equation (4):

$$Tar \rightarrow aCO + bCH_4 + cC \tag{4}$$

This process is essential for proper combustion, in addition to the production of an upgraded and uncontaminated gas that may be sortable to be used in internal combustion engines.

Reduction step is the direct reverse process of combustion, known also as reaction zone, since it involves removing oxygen atoms from combustion products, under high temperature conditions (650–900°C) resulting in obtaining flammable gases. In the gasifier, reduction takes place when CO_2 or water vapor streams pass over hot charcoal containing carbon that is known for its high reactivity with O_2 (Farzad, Mandegari, and Görgens 2016). Thus, oxygen molecules are removed from both CO_2 and water vapor, causing rearrangement of the locations of the single bonds within the particles. During this process, CO_2 is converted to CO molecules, and water vapor is converted to H_2 and CO in which are both flammable gases, according to equations (5–9) (Rajvanshi 1986):

$$C + CO_2 \rightarrow 2CO \; \Delta H = -\ 164.9 \frac{MJ}{kg} mole \tag{5}$$

$$C + H_2O \rightarrow CO + H_2 \;\; \Delta H = -122.6 \frac{MJ}{kg} mole \tag{6}$$

$$CO + H_2O \rightarrow CO + H_2 \; \Delta H = +42 \; \frac{MJ}{kg} mole \tag{7}$$

$$C + 2H_2 \rightarrow CH_4 \; \Delta H = +75 \frac{MJ}{kg} mole \tag{8}$$

$$CO_2 + H_2 \rightarrow CO + H_2O \; \Delta H = -42.3 \frac{MJ}{kg} mole \tag{9}$$

Figure 11 illustrates the following main types of the gasifiers: updraft, downdraft, fluidized-bed and entrained flow.

In an updraft gasification system, the biomass feed enters from the upper edge of the reactor, while the air, oxygen, and steam streams enter from the bottom of the reactor, in a counter-current manner. On the other hand, downdraft gasifiers are designed in a co-current manner, in which both the biomass feed and the gasification material flow along the same route. For a fluidized-bed gasifier, energy and species are transferred by an inert or a catalytic agent. As for entrained flow gasifiers, the biomass is present in dust form, where the raw material and air streams are transported co-currently, and the chemical reactions take place at a temperature range of 1300–1500°C, forming a thick cloud of particles (Corella, Toledo, and Molina 2008). Finally, another type of gasification is the plasma gasification system, where plasma serves as a heat source to enhance gasification and tar cracking.

The most important product obtained from biomass gasification is the syngas, where its composition is determined by identifying the gasifying medium (Corella, Toledo, and Molina 2008). In case of low temperature gasification, heavier hydrocarbons are also produced along with CO and H_2, therefore, the

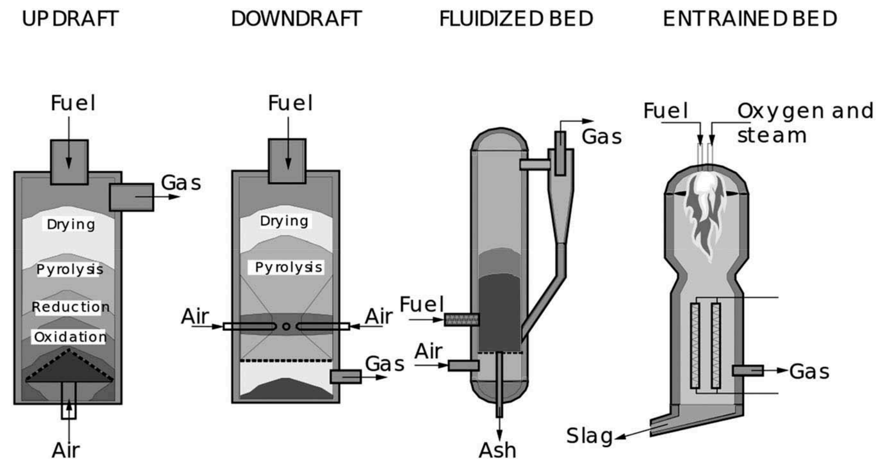

Figure 11. Types of gasifiers (Yin et al., 2004).

gasification temperature should be increased in order to obtain maximum syngas yield along with the least tar formation. Extending the volatile residence time often increases tar cracking. Biomass gasification often involves a high alkali content, which affects the softening temperature of the generated ash. This may consequently cause agglomeration problems, and limit the maximum allowable gasification temperature (Sikarwar et al. 2016). Since a high ash content causes slagging, biomass with a low ash content is favored for syngas production (Oladeji, Itabiyi, and Okekunle 2015). Considering the requirements of the downstream process, gasification is often followed by the shift reaction to adjust H_2/CO ratio.

Syngas may directly be transformed into liquid hydrocarbons, such as diesel and kerosene fuels. Moreover, syngas can also be transformed into other forms of gas fuels, by various processes such as Fischer–Tropsch synthesis, mixed alcohol synthesis and methanol to gasoline process. The options available for syngas conversion are many, and lead to a range of biofuels that contain bio-alcohols such as methanol, ethanol and liquefied gas.

The quality of the product resulting from biomass gasification is greatly influenced by different factors, such as the feedstock type and the moisture content. Cellulose, hemicellulose, and lignin are the major constituents of biomass. Hence, the amount of cellulose and hemicellulose determines the yield of the biogas. On the other hand, the lignin content often determines the amount of oil in the product. The feed moisture content also affects the exit gas properties, and the optimal operating conditions of the gasifier. For instance, the partial pressure of the gasifying agent greatly impacts the reactivity of biomass char, while the temperature and heating rate lead to a higher gas and lower tar production (Oladeji, Itabiyi, and Okekunle 2015). Moreover, it was found that decreasing the particle size also enhances syngas efficiency, and decreases tar yield. Finally, a higher ER (equivalency ratio) results in a lower H_2 and CO yield, and a higher CO_2 content, thus decreasing the calorific content of the product gas (Oladeji, Itabiyi, and Okekunle 2015).

12. Pyrolysis

As mentioned earlier, pyrolysis is thermal breakdown of the materials without air. Pyrolysis mainly converts biomass into bio-solid (coal), bio-liquid (bio-oil) and biogas. A typical process consists of three main phases. The first phase, known as pre-pyrolysis, occurs at temperatures varying from 120°C to 200°C (Corella, 2008). Pre-pyrolysis involves bond breaking, free radicals appearing and carbonyl group formation. The second phase is the main contributing stage, which involves solid breakdown. The third and final phase is continuous char devolatization, which takes place due to the splitting of C-H and C-O bonds (Corella, Toledo, and Molina 2008).

Pyrolysis can be further classified into slow, fast, and flash. Each differs in terms of the heating rate. For instance, in slow pyrolysis, biomass is heated up to 10–20°C/min (Corella, Toledo, and Molina 2008). In fast pyrolysis, biomass is heated at high heating rates, in the absence of oxygen, while in flash pyrolysis biomass moves rapidly along a hot tube by gravity or in a gas flow, using high temperatures and short residence time (Corella, Toledo, and Molina 2008).

The products of pyrolysis are char, pyrolysis liquid, and gas. Char is the solid residue after pyrolysis, and is believed to be the key factor for polycyclic aromatic hydrocarbon (PAHs) formation during biomass pyrolysis, mostly at low temperatures (Corella, Toledo, and Molina 2008). Char can be upgraded into activated carbon and used in purification processes. The liquid product from biomass pyrolysis is known as bio-oil, can be obtained at short residence times, coupled with rapid cooling system to obtain it in a condensed form (Corella, Toledo, and Molina 2008). Usually, this condensate is not at its thermodynamic equilibrium under storage temperature conditions; hence, its chemical composition tends to change toward thermodynamic equilibrium during storage. Finally, the produced gas consists of H_2, CO_2, CO, and CH_4 with traces of C_2 species. CO_2 and CO are created at low temperatures, while H_2 is created at high temperatures. The quantity and quality of the pyrolysis yields are mainly determined by the physical and chemical characteristics of the biomass raw material such as the particle size, elemental composition, temperature, pressure, retention time, and heating rate (Corella, Toledo, and Molina 2008).

At low pyrolysis temperatures, usually below 150°C, tar formation does not take place. However, as the temperature starts to increase, the formation of char begins to take place, resulting in higher yields of gas and lower yields of char (Putun, Uzun, and Putun 2007; Fapetu 1994; Zanzi et al., 2002). Temperature also affects the product composition. Studies by Zanzi, et al. (2002) and Della Rocca et al. (1999) show that the carbon content in the pyrolysis products increases with increasing temperature, while the hydrogen and oxygen contents decrease.

The type of biomass feedstock also affects the composition of pyrolysis products obtained, since there can be great variations in the physical and chemical structure of different feedstock, as well as in the composition of different biomass materials. For example, hardwoods yield lower char products compared to soft woods, while agricultural crop residues produce more char and less gas yields than woods. Zanzi et al. (2002) conducted rapid pyrolysis on wood, olive waste, and wheat straw, and showed that wood gave a more volatile yield, and less char than both agricultural residues. Their findings may be attributed to the higher char yield for the agricultural residues which favors the charring reaction. Their findings also showed a higher yield for olive waste compared to wheat straw, due to the higher lignin content in olive waste.

Moreover, pyrolysis product yields including char, liquid, and gas depend on the applied heating rate to a certain extent. For example, Weerachanchai, Horio, and Tangsathitkulchai (2009) studied various heating rate effects on the yields of pyrolysis of palm kernel cake and cassava pulp residue where heating rates varying between 5°C and 20°C/min were considered at pyrolysis temperature of 700°C. It was found that the attained yield of pyrolysis products are not significantly affected over this relatively narrow heating range. Those results agree with the work by Putun, Uzun, and Putun (2007), which concluded that pyrolysis at temperature range of 500°C to 700°C did not have any effect on the products using a heating rate of 7°C to 40°C. On the other hand, Karaosmanoglu et al. (1999) showed that heating rates more than 30°C/min result in greater yields of liquid and gas, and lower yields of char than those derived from pyrolysis at 10°C/min.

Retention time, also known as residence time, is defined as the time pyrolysis products remain in the reactor chamber before they are discharged out of the reactor. According to Zanzi et al. (2002), the retention time greatly affects the products attained from pyrolysis. An increase in retention time may lead to secondary reactions, thereby resulting in secondary products. Aquino (2007) varied the retention time in batch pyrolysis of cotton gin trash, and found that the char yield slightly drops, while the gas yield increases with increasing retention time. Moreover, Gan and Juan (2008) determined an optimal retention time for bio-oil yield. The oil yield was found to reach a maximum of 21 w.t%, after 20 min, and was then found to decrease when a retention time longer than 20 min is applied.

The particle size distribution and moisture content also affect the properties of the pyrolysis products. The specific surface area of particles or pieces undergoing a pyrolytic process greatly affects the efficiency of the thermo-chemical transformation phase and the reaction time (Zanzi et al., 2002). This was also concluded by Zanzi et al. (2002), which considered the fast pyrolysis of wood and agricultural residues in a reactor that depends on gravity. The char yield was noticed to drop, whereas the gas yield was noticed to increase when using smaller particles of wood and olive waste. In a comparable experiment done by Zanzi et al. (2002), the char yield attained from the pyrolysis of pelletized wheat straw was higher than that attained from untreated straw. Weerachanchai, Horio, and Tangsathitkulchai (2009) studied the change in the yields when using particle sizes of palm kernel and cassava pulp residues of sizes that vary from 0.71 to 3.56 mm at 700°C. Their results showed that pyrolysis of both types of biomass with an average particle size of 2.03 mm caused the highest liquid yield, with 54.3 wt% obtained from palm kernel and 42.4 wt% from cassava pulp residue. In addition, increased moisture content was noticed to steadily increase char yields from pyrolysis (Zanzi et al., 2002). Bridgewater and Peacocke (2000) report that fast pyrolysis involves a properly dry raw material, with about 10% moisture, so that the moisture does not affect the ratio of temperature increase. However, slow pyrolysis can accept higher moisture content levels.

13. Combustion

The final form of biomass thermal use is combustion, also known as oxidation. Combustion is a series of complex consecutive homogeneous and heterogeneous reactions that require a fuel source, an oxidizing agent (usually air) and a heat source. The main yields from combustion are CO_2 and water. At a temperature close to the temperature of the combustion chamber, the biofuel particles entering the combustion system heat up. During the heating process, the particles dry up due to the evaporation of water present in the pores of the elements, since humidity is found at the surface of the particles. In the devolatization step, the degradation of the carbonaceous elements takes place under the effect of heat, where the structure and quantity of the volatile elements produced at this phase are affected by the heating rate of the particles as shown in Figure 12. These mechanisms can be investigated through thermogravimetric analysis (TGA). For example, in the work of Abu Tarboush et al. (2015), thermogravimetric analysis was used in order to investigate the oxidation process of asphlatenes, so that the amounts of asphlatenes adsorbed can be found from the mass loss from the results obtained via TGA.

As for particle fragmentation, this takes place simultaneously with the thermal breakdown of the biomass particles, and is caused by its sudden exposure to heat, known as thermal shock, and the aeration and production of volatile matter. However, abrasion is caused by the impacts that occur between the particles and the sides of the combustion chamber. The combustion process also involves the reaction of the volatile elements emitted during the devolatization phase with O_2. Oxidation takes place heterogeneously, in three distinctive mechanisms as shown in Table 4.

A comparison between the three modes of thermal conversion is necessary. Such comparison can be made at many levels (e.g. required products and their use, technology development, economic feasibility, etc.,.) (www.suncat.ac.uk). For example, if a heating duty is required for processes such as water heating or space heating, oxidation has been found to be the most attractive amongst all options, as well as the most economically feasible.

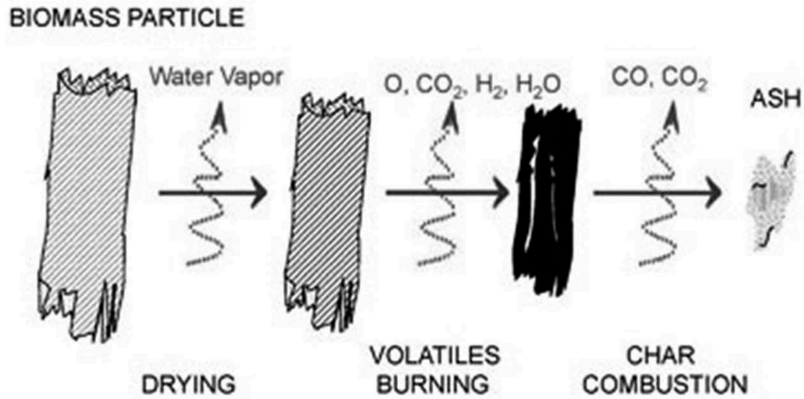

Figure 12. Stages of combustion of biomass particles (adapted from the Canadian Biomass Magazine).

Table 4. Oxidation process mechanisms.

Mechanism	Description
Shrinking Particle Model	the particle is oxidized externally, where the density stays constant, but the particles start to shrink down
Constant size	the particles start to oxidize internally through their pores, where their initial size is preserved by reducing the density
Shrinking Core Model	the particles are now completely oxidized, and since oxidation occurred from the outer to inner surface, ash rounds the combusted zone, but the particles maintain their initial size the same

However, if char, oil, and gas are desired products, gasification or pyrolysis conversion schemes processes should be utilized. Studies reported that gasification was found to be the most efficient process to obtain an enriched hydrogen gas stream (Inayat et al., 2010; Rupesh, Muraleedharan, and Arun 2014; Wu 2013). The enriched hydrogen stream can be further utilized for power and electricity generation, when integrated within gas turbines or fuel cells, thus boosting the systems' efficiency. Additionally, when the obtained gas is purified and treated, it can be used as a raw material in many chemical industries such as catalytic methanol production (www.suncat.ac.uk). Both pyrolysis and gasification processes produce bio-oil which is considered a fuel source for boilers and engines, which can be further treated and used as a fuel source for transportation. Bio-oil is more easily transported than hydrogen gas; however, storage issues are often associated with bio-oil since it is a highly corrosive substance (Zanzi et al., 2002). Additionally, both gasification and pyrolysis processes produce char as a by-product, but slow pyrolysis generates more char than gasification (Roos 2010). Char can be used for soil enrichment, and as a precursor to activated carbon (Roos 2010).

In terms of the level of technology, oxidation is a very well-known technology which has been used for many decades (Roos 2010). On the other hand, pyrolysis is a new technology that is not as widely used as oxidation and gasification systems. Moreover, pyrolysis reactors are still not well-developed, compared to gasifiers. Since gasification is considered a mature technology, many projects are being built using gasification systems. However, gasification processes still face some challenges, especially tar removal options from the obtained gas, which still requires further investigation.

Table 5 summarizes the main aspects of the three modes, including the products obtained, the High Heating Value (HHV), the oxidant used in the process, the typical operating temperatures, the pollutants produced, and the process efficiency, defined as the energy obtained from the process over the ideal energy that could be obtained if the thermal conversion was complete.

14. Environmental assessment of the pelletization process

In order to assess whether biomass pellets can be considered an environmentally efficient energy source, the amount of greenhouse gas emissions that are generated via the different biomass burning techniques need to be evaluated. Different methods may be used to evaluate potential environmental impacts such as the Canadian Environmental Assessment Act (CEAA), the New Brunswick Environmental Impact Assessment Regulation (the "EIA Regulation"), and the Life

Table 5. Comparison of the 3 modes of thermal conversion (adapted and modified from Rupesh, 2014).

Process	Gasification	Pyrolysis	Combustion
Products	With air: CO_2, H_2O and N_2 gases, heat, tar and combustible gases: CO, H_2 and CH_4	oil, tar, CO_2,H_2O, combustible gases: CO, H_2, CH_4 and char	heat, CO_2, H_2O, N_2 gases
High Heating Value (HHV) MJ/kg	5–20	16–19	15–29
Oxidant	air, pure oxygen, steam or their combinations.	-	air
Operating Temperature (°C)	550–900	500–800	850–1200
Pollutants	particulates, tars and compounds of chloride, nitrogen and sulfur	particulates, tars and compounds of chloride, nitrogen and sulfur	particulates and compounds of chloride, nitrogen and sulfur
Efficiency (%)	50–60	45	65

Cycle Assessment Method. The life cycle analysis (LCA) it is a general methodology to evaluate the "cradle-to-grave" energy and the ecological effect of a process or product, and is greatly used as an evaluation method for many systems (Yin 2004).

Life Cycle Assessments (LCAs) study the possible environmental influences over a product's or system's lifecycle starting from raw material collection, all the way through production, utilization and even disposal (ISO 2006). The scope of conducting a LCA is to make a complete analysis regarding the releases and resource necessities of a certain product or system. Hence, all the stages that are involved in the collection, purifying, transportation, and usage of materials must be considered.

The International Standards Organization (ISO) has published a series of LCA standards. According to the ISO 14,040, a systematic LCA analysis has the following structure, which is also summarized in Figure 13.

(1) Goal and scope definition: It starts with defining and describing the objective of the analysis, discussing any assumptions and statistics, specify the system boundaries and the environmental influences to be considered.
(2) Life Cycle Inventory (LCI): It is compiling data on energy, material currents, and emissions. The result of this phase is an inventory of all contributions and productions in the form of fundamental runs to and from the environment for all the procedures the study includes.
(3) Life Cycle Impact Assessment (LCIA): It discusses the effects associated with the service under study, are evaluates the process in terms of impact categories. Various influence classes can be joined together using weighting to contribute to the complete impact assessment.
(4) Interpretation: results from the preceding steps may be explained in terms of the objectives of the study.

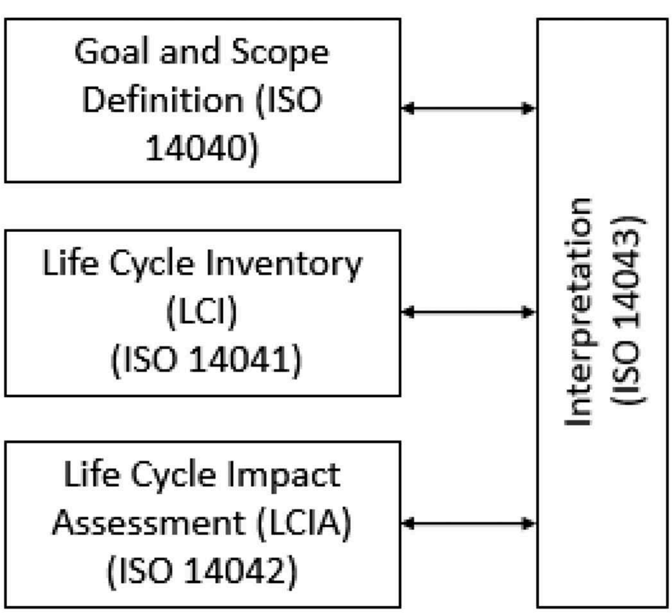

Figure 13. Outline of LCA Stages with the Used Standards.

Various researchers have studied GHG emissions from different types of biomass, or feedstock used as raw materials in energy production. Comparative LCA study has been recently conducted to compare two different gasification systems (downdraft gasifier and CFB gasifier). H_2 production proved that downdraft gasifier delivered better environmental performance over CFB gasifier (Susmozas et al. 2016). In addition, Susmozas et al. (2016) conducted another LCA study of hydrogen production, they focused on direct emission, outer power production, and biomass manufacture to be major practices in determining the environmental impacts, and it has been proved that bio-hydrogen production with CO_2 capture delivers better environmental performance over conventional processes. Another study proved electricity production from biomass delivered noticeably less CO_2 emissions than coal fired systems (Cambero, Alexandre, and Sowlati 2015; Varun and Bhat 2009b).

Life cycle analysis for GHG emissions resulting from bioenergy systems in British Columbia was investigated by Cambero, Alexandre, and Sowlati (2015). The results showed communities in which all energy needs were met only using fossil fuels and biomass residues burnt off, a net reduction of up to 40,909 ton of CO_2 equivalent GHG emissions could be achieved. In case the energy needs can be met through renewable sources, the net achievable GHG emissions reduction was significantly lower. A LCA study was conducted for a pelleting process that uses waste olive husk in Cyprus, and the results from this analysis indicate that when renewable energy systems were applied in the manufacturing stage, the environmental effects of producing olive husk pellets ware enhanced by more than 85%. Farzad, Mandegari, and Görgens (2016) presented a literature review that summarizes different environmental assessments conducted for gasification, based on feedstock, technology, and product as shown in Table 6.

15. Conclusion

This review paper discusses the main factors that affect the pelletization process of biomass raw materials. An overview of many different types of biomass raw materials, which may be used for pelletization, have been presented. Moreover, each of the different stages that make up the pelletization process has been discussed. It was found very essential to optimize the design and operating conditions of the system, to be able to synthesize pellets with high durability. For instance, the feed moisture content was found one of the key factors that affect pellet durability. Most studies recommended an optimal moisture content value is between 8% and 12%, to attain the best pellet durability values. Also, optimum particle size distributions of the feed material that help to attain the best pellet durability and bulk density values, have been presented. Some studies revealed that the recommended values for moisture content and particle size distributions are subject to change, since many other factors interfere with the pelletization process, such as heat pretreatment and the type of the biomass raw material itself regarding the moisture content.

Table 6. Environmental assessment on gasification (Farzad S, 2016).

Purpose	Case	Scope	Reference
Evaluation of feedstocks	Evaluation of co-firing of biomass with coal Alternatives:Wood pellets, coal, and natural gas	GHG emissions	Zhang, 2009
	Evaluation of co-firing of biomass with coal for electricity production *Alternatives*: Forestry residues, energy crops, and coal	GHG emissions	Froese, 2010
	Heat production through gasification *Alternatives*: Forestry residues, and recycled wood	GHG emissions	Puy, 2010
	Heat production through gasification *Alternatives*: Forestry residues, wood pellets, and natural gas	GHG emissions	Pa, 2011
	Heat production through gasification *Alternatives*: Forestry residues, woody energy crops, and natural gas	GHG emissions	Pucker, 2012
	Production of H_2 through gasification *Alternatives*: Vine and almond pruning, forest waste from pine, and eucalyptus plantation	FU*: production of 1 Nm^3 H_2	Moreno, 2013
Evaluation of technologies	Evaluation of integrated gasification combined cycle (IGCC) *Alternatives*: IGCC with upstream CO_2 adsorption *vs.* chemical absorption of CO_2	FU: produced energy unit	Carpentieri, 2005
	Evaluation of H_2 production via biomass gasification *Alternatives*: Gasification followed by syngas reforming *vs.* electricity generation	GHG emissions	Koroneos, 2008
	Evaluation of production processes for ethanol production *Alternatives*: Biochemical *vs.* thermochemical processes	GHG emissions	Bright, 2009
	Evaluation of CHP plant with different sizes *Alternatives*: 0.1, 1, and 50 MWe	GHG emissions	Guest,2011; Kimming,2011
	Evaluation of CHP plants for power and heat production in rural areas *Alternatives*: Biomass fed CHP *vs.* fossil fuels in a large scale plant	FU: 1 year supply of heat and power to a modern village	Kimming, 2011
	Comparison of different gasifiers for H_2 production *Alternatives*: Downdraft gasifier and fluidized bed gasifier	GHG emissions	Kalinci, 2012
	Evaluation of energy production systems *Alternatives*: Electricity *via* gasification *vs.* bioethanol through enzymatic hydrolysis	FU: the use of biomass chips from 1 ha	Susmozas, 2016
	Evaluation of potential future energy systems *Alternatives*: F-T liquid through biomass gasification, rapeseed based biodiesel, and fossil fuels	FU: 1 energy unit of diesel fuel	Tonini,2012
	Evaluation of methanol production (*via* gasification) based on an autonomous distillery or sugar mill *Alternatives*: Co-generation plant combined with methanol or biomass integrated gasification/gas turbine (BIG-GT) system	1 MJ of each product	Renó, 2014
	Evaluation of bioenergy generation alternatives using forest and wood residues *Alternatives*: Combustion and gasification technologies with different capacities	GHG emissions	Cambero, 2015
	Evaluation of H_2 production through biomass gasification *Alternatives*: Bio-H_2 with/without CO2 capturing *vs.* fossil based H_2	FU: 1 kg H_2 produced	Susmozas, 2016
Product alternatives	Evaluation of synthetic natural gas (SNG) production through gasification *Alternatives*: SNG for heat, power, and transportation	GHG emissions	Steubing, 2011

* FU; Functional unit of LCA study

Compressive forces were recommended to be taken into consideration rather than the tensile ones, since they are more involved in the packing and transportation process.

The use of binders that enhance bonding within the pellet molecules have also been reviewed, together with the different binding mechanisms. In terms of the types of binders to be used, many of the studies concluded that sodium silicate is highly utilized as a binder, since it is very effective when it comes to enhancing the pellet compressive strength. Other types of binders may also be used such as calcium lignosulfonate, lithium silicate, lignosulphonate, corn starch, and bentonite, which are effective for enhancing the binding properties within the particles of the pellets and consequently enhancing the final pellet quality. A new parameter (Q) was proposed to be taken into consideration rather than the durability only, which counts for all the affecting parameters on the pellets quality throughout the entire process.

Additionally, the consequence of using feedstock with unlike properties, on the properties of produced pellets have been reviewed. Moreover, other parameters that affect the pelletization process, such as the forming pressure, pellet mill variables, and roll press variables have been presented. Finally, post pelletization techniques have been discussed, mainly gasification, pyrolysis and combustion, and a life cycle assessment for a pelletization process was briefly presented, which mainly discussed the environmental impacts of this process from the very beginning to the very end.

Acknowledgments

The authors would like to thank the financial support of the Munib and Angela Masri Institute of Energy and Natural Resources at the American University of Beirut.

Nomenclature

BD	bulk Density
C	carbon
d.b	dry basis
Du	durability
FC	fixed Carbon
GHG	greenhouse gases
H	hydrogen
HC	hydrocarbons
LCA	life cycle assessment
LHV	lower heating value
MC	moisture content
N	nitrogen
NOx	nitrogen oxides
O	oxygen

OL	olive leaves
OP	olive pruning
OW	olive wood
PA	proximate analysis
Pa	pascal
PD	particle Density
Q	quality
S	sulphur
UA	ultimate analysis
VM	volatile Matter
VOC	volatile organic compounds
w.b	wet basis
wt	weight

Funding

This work was supported by the Munib and Angela Masri Institute of Energy and Natural Resources.

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