Effect of binders on agricultural crop residues and wastes pellets. A review.

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ABSTRACT Agricultural crop residues and wastes can be pelletized to improve their undesirable characteristics such as low bulk density, poor transportation and storage properties, and low volumetric energy. Trading of agricultural crop residue and wood pellets within North America and Europe has seen a significant growth in the past few years. This is because of potential utilization and application in heat, power and combined cycle generation, gradually replacing fossil fuels. Binders in pellets are employed to improve binding, lubrication, and abrasion characteristics by forming matrix which improves intermolecular bonding within the contact area of the biomass during pelleting. The aim of the study is to review the effect of different binders on the physical properties of agricultural crop residues and wastes pellets. The cost of binder addition during pelleting is reviewed, a sensitivity factor in production cost of a pellet.

Keywords: Densification, Biomass, Pretreatment, Binder, Cost evaluation.
INTRODUCTION

Increasing demand for global energy is taking a shift to renewable, non-fossil sources with low carbon footprint (Whalen et al. 2017) and the continual energy dependency have been the motivation for increased used of bioenergy as an alternative in both transportation sector and electricity generation (Handra and Hafni 2017). Increasing population and living standards contribute to drive excessive consumption of fossil fuel, resulting in high levels of environmental pollution (Chen et al. 2015). Shifting to renewable energy sources like biomass could reduce the amount of greenhouse gases (GHG) produced (Emadi et al. 2017). Considerable research is ongoing on the potential biomass energy supplies, land use change and GHG emissions to ascertain sustainability (Muth et al. 2013; Ebadian et al. 2012). According to the U.S Energy Efficiency and Renewable Energy (EERE 2012), biomass utilization as renewable energy source will reduce dependence on crude oil, promote diverse use of domestic and sustainable energy resources, and reduce carbon emissions from energy production and consumption.

Different types of feedstocks such as agricultural crop residues, municipal solid waste, industrial and domestic waste, and micro-algae are promising sources of energy, and can produce biofuels through biochemical and thermochemical conversion. Lignocellulosic biomass from agricultural crop residue origin is destined for direct combustion and thermochemical conversion during densification to improve storability, handling and transportation thereby increasing the energy density (Whalen et al. 2017).

Lignocellulosic biomass is composed of cellulose, hemicellulose and lignin, organic extractives, and inorganic minerals. Cellulose, hemicellulose, and lignin are the main components of biomass and their weight percentages depend on the type of biomass (Chen 2015). Cellulose contains crystalline and amorphous structures while hemicellulose has a branched mixed polymerized monosaccharide. Lignin is a three-dimensional structure that is highly branched and contains polyphenolic substance (Balat et al. 2008; Aho et al. 2013). Therefore, an effective pretreatment technique is required for depolymerization of lignin and disintegration of the recalcitrant structure to open channels for cellulose and hemicellulose conversion reactions (Agu et al. 2017; Chaturvedi and Verma 2013). Different pretreatment techniques of biomass show various reactivity to lignocellulosics for energy utilization.

Densification of biomass is accomplished by applying mechanical force to compact biomass into uniformly sized solid particles such as pellet, briquette, log, and cubes (Veal 2010; Chen et al. 2015). Pelletization process shows high productivity, density and strength and pellets formed from herbaceous biomass do not have enough strength due to lack of natural binding components between the particles. Utilization of such pellets will be expensive, dusty and difficult to handle (Lu et al. 2013). Therefore, agricultural crop residues when ground and after physico-chemical and biological pretreatment could be compressed or densified using different chemicals, additives or binders to enhance pellet durability and strength. A good number of studies using different pretreatment methods and binders have been conducted to address pellet quality using various feedstocks (Lu et al. 2013; Agu et al. 2017; Emadi et al. 2017; Soleimani et al. 2017; Tilay et al. 2015; Kaliyan and Morey 2006). According to Emadi et al. (2017), the use of binder additives boosts binding through the interlocking of biomass particles and, improves the pellet quality produced. Generally, densification of herbaceous biomass depends on strength and durability of the particle bonds and these are the main desirable parameters use to describe biomass pellets (Tumuluru et al. 2011; Agu et al. 2017).

A binder can be in liquid or solid form to facilitate strong inter-particle bonding. Many types of binders have been investigated and applied in different densification of biomass. During binding reactivity, preheating or steam conditioning is important to add heat and moisture to activate the added binder; the choice of binder depends on cost and environmental sustainability (Kaliyan and Morey 2006). Also, the selection of binder additive in any process is dependent on the binder-temperature reaction (Yoo and Jo 2003). Soleimani et al. (2017) reported that the binding capacity
of herbaceous biomass can be improved by the application of pre-processing method or direct application of binders. Tumuluru et al. (2011) reported that binders reduce the wear on production equipment and increase the abrasion resistance of the fuel. Many research studies have reported many binders or additives which are commonly used for densification using different biomass (Pietsch 2002; Obernberger and Thek 2010; Tarasov et al. 2013; Sokhansanj et al. 2005; Kaliyan and Morey 2006). According to Tarasov et al. (2013) on EU standard, binder additives that improve fuel quality, decrease emissions or improve burning efficiency constitute a maximum of 2% of the total mass of the biomass pellets.

Binders can be categorized into three types: a) organic binders such as molasses, corn or potato flour, maize and rye flour; biosolid, microalgae, walnut and peanut shell, woody plants, coffee meal, bark, hydrothermal carbonize biochar, softwood residue, sawdust; b) inorganic binders include lignosulfonate, recycled polymer plastic, bentonite, colloids, lubricant, sodium hydroxide, calcium hydroxide, potassium hydroxide, dolomite, lime, cement and alkaline lignin; and c) complex binder like coal tar residue (Soleimani et al. 2017; Yoo and Jo 2003; Lu et al. 2013; Kaliyan and Morey 2006; Emadi et al. 2017). Organic binders can be classified as biological additives which are rich in starch content, free of ash and unwanted elements like nitrogen and sulfur whose two elements can be sources of gas emissions. Carbohydrate as a binder in biomass pellets would lower ash content, compatible with the pellet structure, and as a stabilization aid whereas the disadvantage when used in thermochemical processing relates to generation of pollutant gases which are not friendly to the environment (Soleimani et al. 2017; Yoo and Jo 2003). Whittaker and Shield (2017) reported that starch and water-soluble carbohydrates improve biomass pellet durability and organic binders are employed to improve the strength of the ceramic pellets or increase the porosity of the final product after combustion during thermal processing. Inorganic and complex binders are referred to as commercial binders, rich in minerals like lignosulfonate (water-soluble anionic polyelectrolyte polymer) or bentonite (clay) or coal tar residue (coal). The main advantage of using inorganic binders in biomass pellet is to achieve satisfactory mechanical strength (Kaliyan and Morey 2006; Tarasov 2013). Complex binder can improve the mechanical strength and calorific value of pellets, and reduces energy consumption involved in the granulation. The disadvantage is on environmental pollution and there is insufficiency research study on the pollutant emission performance of biomass pellet with coal tar residue as a binder (Si et al. 2017). In general, the use of binders in biomass pellets for bioenergy is allowed but needs to be specified as part of the pellet material constituent (Tumuluru et al. 2011). Binder selection influences the cost of pellet production and many research studies have investigated and evaluated various binders used in pellet production using different pretreatment techniques to support their results (Pirraglia et al. 2013). There is no literature on the cost of binder which is part of variables that influence pellet characteristics, economics, and storage properties.

Therefore, the purpose of this review study is to evaluate the effect of different binders on the physical properties of agricultural crop residues and wastes pellets, and detailed studies in the addition of binders using various pretreatment processes and densification of biomass. Emphasis is also placed on the cost of binder, a sensitivity factor in pellet production facility evaluation.

Densification

Densification process may be achieved through pelletization (Veal 2010). The aims of densification are to increase bulk density from 40 – 200 kg/m³ to final density of 600 – 1400 kg/m³, to improve storability, lower down handling and transportation cost, and lowering of moisture content of biomass (Mani et al. 2003; Chen et al. 2015). Dueck et al. (2017) reported energy values of densified biomass suitable for use as a fuel source and these include: ease Strength, high calorific value, high content of volatiles, high combustion rate, and low activation temperature. Strength and durability of densified biomass are influenced by binders, die diameter
and temperature, pressure and pre-heating of biomass mix (Tumuluru et al. 2011). In addition, densification process is affected by physical and chemical characteristics, binders and biomass source (Dueck et al. 2017). However, this review will focus more on the binder effect in describing the biomass pellet strength and durability. Advantages of binders in pellets are to improve binding, lubrication, combustion properties, and abrasion characteristics, and assist in forming matrix, film of chemical reaction that strengthens interparticle bonding (Tumuluru et al. 2011; Pirraglia et al. 2013).

**Biomass Binding Mechanism During Densification**

Biomass binding forces during pelletization can be used to measure the strength and durability of densified biomass which depends on physical forces that bond the particles together (Kaliyan and Morey 2006). Pietsch (2002) and Kaliyan and Morey (2006) reported that there are five main groups of binding forces that act within the individual particles of densified products. These binding forces have been observed in different densification process via pharmaceutical powders, animal feeds and biomass materials. These binding forces are (i) solid bridges: formed between particles due to crystallization of some molecular components, chemical reaction, hardening of binders and solidification of melted components, and are mainly formed after cooling of pellets; (ii) Attraction forces within solid particles of pellet: these are short-range of attracting forces which cause solid particles to bond together within the close matrix and such forces are molecular, hydrogen, electrostatic, and magnetic forces; (iii) mechanical interlocking bonds: this is bonding together of fibers and particles during compression process; (iv) adhesion and cohesion forces: these are high viscous binders which adhere to the solid particles surfaces to produce strong bonds very similar to solid bridges in the bonding mechanism; and (v) interfacial forces and capillary pressure: free moisture between particles in a wet agglomeration process causes bond cohesive forces generated from interfacial tension at the liquid-gas interface. Interfacial forces and capillary pressure bonds generated during agglomeration disappear immediately the free moisture evaporates and possibly some other binding mechanisms may take over.

**Effect of Binders on Pellet Quality**

**Densification Measuring Variables**

The quality of biomass pellet produced after densification process can be measured in terms of strength and durability, dimensional stability, and pellet density. The effectiveness of the inter-particle bonds generated during densification process is used to measure these variables in describing pellet quality. Pellet quality is evaluated by considering the level of importance. Pellet strength and durability are the most important property due to the physical resistance of pellets to forces in pellet handling and transportation. This is followed by dimensional stability and pellet density indicating less dust generation during handling and storage (Agu et al. 2017). However, when these variables values of pellets do not match with the quality or market standard, a range of additives (0.5 to 5% by weight) are added to the biomass either to increase or minimize the pellet quality (Tabil 1996). Traasov et al. (2013) reported that Europe (EU) and United States of America have standards for additives use in improving fuel quality, decrease emissions, or boost burning efficiency. However, some EU countries prohibits addition of binders in pellets whereas in Austria, especially biological additives, only 2% (by weight) are allowed for wood pellet production (Obernberger and Thek 2004).

**Pellet Strength and Durability**

Pellet strength or compressive resistance is the maximum crushing load a pellet can withstand before cracking or breaking (Kaliyan and Morey 2006). Pellet strength is determined by a diametrical compression test using tensile strength calculation based on facture load (Tabil 1996; Fell and Newton 1968 and 1970). Durability is defined as abrasion resistance due to impact
resistance and shearing of pellets over each other and over the wall of the tumbling can, and this can be done using ASABE 269.4 Standards (2003) (Kaliyan and Morey 2006; Tarasov et al. 2013). Tarasov et al. (2013) reported that pellet durability of EU standards should not be less than 97.5% and stable with moisture content values ranging between 9% and 14% wb.

**Pellet Density and Dimensional Stability**

Pellet density is calculated by measuring the length, diameter and mass of the pellet immediately after ejection, sealed in Ziploc bags and stored at ambient temperature for 14 days. The dimensional stability of the pellet is calculated based on pellet changes (after 14 days storage period) in longitudinal and radial dimensions expressed in percentage (Kashaninejad and Tabil 2011).

Emadi et al. (2017) studied the effect of mixing polymer plastic - linear low density polyethylene (LLDPE) (as an additive at four levels 1, 3, 6 and 10%) with torrefied wheat and barley straws and pelletized. They found that adding 10% LLDPE to the torrefied biomass pellets resulted in a 280% and 253% increase in tensile strength for wheat and barley pellets. Adding LLDPE 1% to 10% gave more dimensional stability to wheat straw pellets than barley. Addition of 6% LLDPE to the biomass pelleted to a maximum increased density by 1.8% for wheat and 1.7% for barley. Thus, dimensional stability close to 100% is considered a good result in the case of density.

Agu et al. (2017) reported that mixing alkali solution (NaOH or KOH at two concentrations 0.75 and 1.5% w/v) with canola straw or oat hull and pretreated in microwave oven had a significantly higher pellet density (canola straw 1392.21 kg/m$^3$ and oat hull 1292.59 kg/m$^3$) and tensile strength (canola straw 5.22 MPa and oat hull 3.36 MPa). In other words, increasing the alkali concentration increased pellet density and tensile strength of both samples. The dimensional stability of canola straw and oat hull pellets decreased with increasing alkali concentration. Overall, NaOH pretreated pellets showed better results in pellet quality than KOH pretreated pellets.

Hu et al. (2015) used four different kinds of binders (lignin, starch, calcium hydroxide and sodium hydroxide) to study effects of binders on the properties of bio-char pellets. From the results, they discovered that starch was not suitable for use as binders for bio-char pellets due to low mechanical strength and volume density. NaOH binder showed the highest compressive strength among all pellets, exhibited the highest moisture uptake that may worsen the handling and storage treatment of bio-char pellets. For lignin and calcium hydroxide bio-char pellets enhanced mechanical strength and promoted combustion performance and lower moisture uptake, which demonstrated better potential for utilization as biofuels.

Lu et al. (2014) investigated experimental trials to make wheat straw pellets with wood residue (non-pretreated and pretreated) and binders (5% crude glycerol, 2% bentonite, or 2% lignosulfonate, and pretreated wood residue). The results showed that pellet density significantly increased as a result of adding binders to wheat straw. The binders used significantly increased tensile strength over that of wheat straw without binder with values ranging from 1.13 to 1.63 MPa. The results showed that the binders would work for pelletizing wheat straw as it relates to handling, transportation and storage of fuel pellets.

A study on the quality of oat hull fuel pellets using bio-additives such as lignin and amino acids was reported by Abedi and Dalai (2017). The results showed that oat hull pellets with proline was the best choice as co-binder with lignin. Lignin content ≥ 15% and proline content ≥ 5% had the highest density, durability and hardness.

Peng et al. (2015) used sawdust, starch and lignin as binders for making torrefied pellets. Torrefied sawdust produced from pine sawdust in a fixed bed reactor was mixed with different
binders in a ratio of 5–30% (w/w) and compressed into pellets. Their results showed that torrefied sawdust particles under typical torrefaction conditions (280 – 300 °C for 10 – 30 min) could result in strong pellet and high volumetric density. Slightly higher results were obtained for both starch and lignin binders in torrefied pellet than sawdust as binder, but pellet density is lower compared to sawdust torrefied pellets. They recommended sawdust as an effective low-cost binder choice for densifying torrefied sawdust into torrefied pellets, given its abundant supply and much lower cost compared to starch and lignin.

Tilay et al. (2015) concluded that optimized canola meal pellet with 99% durability and 189 N hardness was produced with 5% (w/w) binder, 2% (w/w) lubricant and 12% (w/w) moisture content. The focus is to pelletize waste canola meal biomass to increase bulk density to reduce cost of transportation and storage and provide better material supply chain in the reactors with less dust formation.

Carbohydrates as binders in biomass densification for biochemical and thermochemical processes was investigated by Soleimani et al. (2017). Carbohydrate (molasses, fructose, maltodextrin, sucrose and glucose) and lignosulfonate binders were applied in two cellulosic biomass, spruce wood shavings and wheat straw. From the results, fructose and molasses worked best as binders for spruce wood shavings and wheat straw. Increased level of binder in each type of biomass increased the durability of the pelletized biomass. Also, the combination of fructose and canola oil was the best selection for spruce wood shavings whereas molasses and crude glycerol were superior selection for wheat straw.

Kashaninejad and Tabil (2011) studied the effect of microwave chemical pretreatment on compression characteristics of biomass grinds. Adding 1 to 2% NaOH or Ca(OH)₂ with wheat straw or barley straw and pretreated in microwave oven followed by densification significantly increased the pellet density in wheat straw and barley straw pellets. The dimensional stability of wheat straw and barley straw pellets decreased with increasing alkali concentration. Increasing the concentration of alkali solution from 1 to 2% significantly increased the tensile strength of barley straw pellets whereas wheat straw pellets had no remarked changes observed in the tensile strength. NaOH improved the pellet density and hardness of barley straw pellet than wheat straw pellet.

Effect of glycerol on densification of agricultural biomass was investigated by Emami et al. (2015). The study used selected agricultural crop residues like wheat, barley, oat and canola straws and mixed with three levels of glycerol (2.5%, 5.0% and 7.5% w/w). The results showed that no-glycerol pellets had higher pellet density than glycerol-added pellets. Canola straw and oat straw pellets had higher pellet density than wheat straw and barley straw pellets. Pellet samples mixed with 7.5% glycerol had a high durability without marked changes in pellet density.

Si et al. (2016) investigated effect of carboxymethyl cellulose (CMC) binder on physical and mechanical characteristics of biomass (cotton stalks, wheat straw and rape straw). The results showed that CMC improved pellet density, compressive strength, and durability for cotton stalks and wheat straw whereas adding CMC decreased rape straw pellet quality. Furthermore, their results indicated that CMC may not be suitable as a binder for densification of biomass with extractives.

**Summary of Observation**

Identifying the effect of binders on pellet density, durability, pellet strength and stability on agricultural crop residues pellets were difficult because different types of binders and pelleting conditions were used in evaluating the pellet quality. Pelleting conditions depend on biomass characteristics and binding mechanism. Across the studies, use of binders improved some factors and temperature of the die low or high enhanced binding ability in evaluating pellet quality. Good
quality pellet can be produced from wood residue, cereal reside and dedicated energy grasses. However, higher quality pellets can be produced from wood-based biomass (Peng et al. 2015). Wood biomass is easier to pellet than cereal residues and this is because cereal residues have lower lignin content and higher extractive content (Whittaker and Shield 2017). In addition, wood-based commercial pellet production is done globally compared to agricultural biomass-based pellets which have limited production (USDA 2009). The cost of some binders was reported but no remarkable amount was quoted to evaluate cost competitiveness. Production cost of pellet is also dependent on binder cost.

Conclusion

Combination of organic and inorganic binders has shown evidence of improved pellet quality especially biomass with lower lignin content. Optimizing the pelleting condition to suit each type of binder to biomass can help improve pellet quality. Therefore, each binder has results in unique physical characteristics when applied with different biomass. Cost of purchase and impact on the environment are important factors in the choice of economics and environmental sustainability.

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