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# Factors affecting wood, energy grass and straw pellet durability – A review



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## ABSTRACT

Pellets produced from wood, energy grasses and straw present a higher energy density feedstock than wood chips or bales, and therefore reduce the costs of handling, transport and storage throughout the supply chain. European specifications provide limits to the proportion of fines (particles less than 3.15 mm) allowed in pellets, which refers to the durability of the pellets. Fines have implications for health and safety in supply chains, and cause issues with slag formation in combustion systems. This paper reviews the factors affecting biomass pellet durability. The industrial trade for wood pellets has expanded greatly over the last decade and involves the international trade of tens of million tonnes annually. Due to increasing demands for pellets, there has been growing interest in utilising more varied biomass types. The aim of this review is to examine feedstock qualities and pelleting conditions that produce durable pellets. Pellet durability can be affected by the feedstock characteristics, the moisture content or size reduction during pre-processing, and by pelleting conditions, can also affect durability, such as the storage conditions and handling frequency, therefore an understanding of all the factors affecting durability throughout the supply chain is needed in order to prioritise where advances can be made.

## 1. Introduction

Pellets are a suitable biomass feedstock for both heat and power applications, with co-firing in coal-fired power stations currently being their main large-scale application. The industrial trade for wood pellets involves the international bulk transport of more than 10 million tonnes annually [1]. The majority of demand for pellets originates from the European Union (EU), in response to its greenhouse gas (GHG) emission mitigation policy [2]. The major import originates from North America but the increasing demands have stimulated advances in Russia, Africa, South America and Asia [1]. Although pellets are more energy intensive to manufacture than wood chips or bales, GHG benefits can still be achieved when using them to displace conventional fossil fuels, even when importing pellets from abroad [3-5]. The trade in wood chips is predominantly limited to between European countries, although there are some instances where longer transoceanic supply chains exist, for example between Japan and Canada [6]. Generally, wood chip transportation into Europe is limited due to necessary compliance with phytosanitary restrictions, which require that imported wood (from specified locations) is treated at 56 °C for 30 min [7,8]. Alternatively chips can be treated by fumigation, in batches smaller than of 2 m<sup>3</sup>, with methyl bromide or sulfuryl fluoride at 80 g/m<sup>3</sup> for more than 24 continuous hours [9]. The practicalities of treating sufficiently large volumes of wood chip in this way, and the bulky nature of the product, mean that now predominantly wood pellets are traded across long distances [6].

In large scale biomass supply chains the increased energy density of pellets reduces costs throughout the supply chain in regards to handling, storage and transport. Pellets are made from dry, untreated, biomass that is hammermilled into fine pieces then reformed into small, cylindrical pellets under high pressure and temperature [10]. Pellets are therefore ideal for co-firing with coal, as they can easily be reduced to dust in coal pulverisation systems and be combusted via direct injection [11]; presenting a relatively inexpensive and easy method of reducing carbon dioxide (CO2) emissions from coal-fired power stations [12]. A complication with this, however, is that their composition means when pellets are either poorly produced, repeatedly handled or stored inappropriately, they break down into smaller particles and fines. Not only does this remove the benefits of having a homogenous and densified fuel, but the presence of fines can have important health and safety implications, and there is even a risk of dust explosions when handling and transporting large quantities of pellets [13]. Also, breakages of pellets can increase losses during the supply chain, which can have negative impacts on the GHG mitigation potential of pelletised fuels [14]. Another important factor is 'customer satisfaction', where it is important to satisfy consumers of pellets who

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demand high quality fuels, and breakages may lead to increasing rejection of pellets and further losses [15].

The content of fine particles in a bulk mass of pellets is extremely dependent on their mechanical durability [16], which can be affected by a number of factors. Generally, pellet production requires a high level of expertise [17]. Pellet suppliers must adhere to strict specifications regarding technical pellet characteristics such durability and fine content (particles less than 3.15 mm), the energy content of the fuel, and ash content. Pellet properties must be monitored because deviations in quality can affect the emissions from combustion and the longevity of combustion systems [16]. For example, the ash melting point affects the temperature at which ashes soften and fuse, and the resulting slag formation can disturb the combustion process by altering primary air flows and overheating the grate. This can also be exacerbated by the presence of fines, which can also cause problems by burning rapidly to generate very high temperatures that can lead to ash melting [18].

The mentioned specifications are set in place in order to regulate the quality of pellets for use in heat and CHP boilers up to 1 MW [19] and there is a separate standard emerging for large industrial applications [20]. A number of national standards exist throughout Europe, but as trade between countries becomes more widespread, it is necessary to harmonise them [16]. The European Standard Committee CEN/TC 335 is expected to overrule other standards describing the technical specifications for all forms of solid biofuel in Europe [21]. The common standard (EN-14961-2) will form the platform for a certification system, identifying the specifications for different categories of pellets [10]. Generally, the highest grades have the strictest standards and offer the best combustion properties. The specifications for heating pellets are stricter than for industrial pellets, requiring lower contents of ash, fines, nitrogen, sulphur and chlorine (Table 1). The IWPB standard also introduces some sustainability criteria to regulate the environmental impacts of sourcing and trading of woody material between countries [1]. Overall, a pellet durability of 97.5% is considered to reach the uppermost standard (EnPlus A1), and the lowest limit, for both industry and domestic use is 96.5%.

Sawdust is an ideal substrate for pelleting as is untreated, and even minor contaminants are removed through bark removal and washing of saw logs prior to sawing. Due to an increasing demand for wood pellets, and a limited supply of sawmill residues, there has been growing interest and exploration in the production of wood pellets from other resources [12]. These include bark, forest residues, cereal residues and energy grasses. As these feedstocks differ in chemical composition, they will undoubtedly produce different qualities of pellets. Some feedstock parameters have a greater effect on pellet durability than others, therefore it is important to understand their importance. In this paper, factors affecting biomass pellet durability are reviewed, including the effect of different biomass properties, how biomass is pre-treated and under what conditions the biomass is pelleted. The objective is to understand the major factors affecting durability throughout the supply chain in order to suggest where advances can be made, and identify where certain feedstocks may or may not be suitable for quality pellet production

## 2. The pelleting process

This report specifically addresses the production of pellets, rather than other densified biomass types such as briquettes or tumble agglomerated products [22]. Pelleting uses a series of rollers to compress biomass through a steel die. In contrast, briquetting produces 'biomass bricks' through compression by rollers rotating in opposite directions [11]. Tumble agglomeration systems involve mixing biomass with binders in a ball drum [22]. Generally, out of these three types, pellets are regarded to be the most durable because they are placed under the highest amount of pressure during formation. Often in the literature the term pellets and briquettes are confused, which is due to them only recently being defined by the European standard EN-14961-2 [12].

Wood pellet mills can reach up to 750,000 t/year [23], whereas straw mills are usually smaller, with a suggested economic optimum of 150,000 t/year [24]. There is high uncertainty over the throughput of pellet mills [5]. On a mill-basis a wood or straw mill would have a typical throughput of 4 and 5 t/h per 250 kW pellet mill, respectively [25], though it can change during the mill's lifetime due to wear [24].

The pelleting process is usually adapted to the specific biomass feedstock, but usually includes the following stages (Fig. 1): reception of raw material, drying, grinding, pelleting, cooling and screening [10]. Initial comminution is performed before drying. Roundwood is typically chipped, waste wood is sorted into grades, tub-ground and screened against plastics and metals (Dalkia Pers. Com. 2013), and bales are shredded. After drying, the material is ground into fine particles using a cutting mill, usually a hammermill. The hammermilled feed enters a mixing chamber where steam and additives are added (Sections 3.3.3 and 3.3.4). The pellet mill consists of a circular die that is perforated with holes that the biomass is forced through via the action of rollers, either by rotating the die or the rollers. The ground biomass is continuously fed into the pellet mill where it is steadily compressed into the pellet channels [26].

## 3. Factors affecting wood pellet durability

### 3.1. Pellet dimensions

The ends of pellets are the main source of fines, as in the pellet die the majority of the heating occurs on the outer sides of the pellet, which

#### Table 1

Current specifications for domestic and industrial biomass pellets [1,19].

			EnPlus Standards			IWPB Standards	
Parameter	Unit	ENPlus-A1	ENPlus-A2	EN-B	I1 Industrial	I2 Industrial	13 Industrial
Diameter	mm		6-8		6 ≤ D ≤ 8	6≤D≤ 10	6≤D≤ 12
Length	mm		$3.15 \le L \le 40$			≤ 40	
Moisture content	% a.r		≤10			≤10	
Ash content	% a.r	≤ 0.7	≤ 1.5	≤ 3	≤ 1	≤ 1.5	≤ 3
Mechanical durability	% a.r	≥ 9	7.6	≥ 96.5	≥ 97.5	≥ 97	≥ 96.5
Fines (<3.15mm)	% a.r	< 1			≤ 4	≤ 5	≤ 6
Net calorific value	MJ/kg a.r	16.5 ≤ CV ≤ 19	16.3 ≤ CV ≤ 19	$16.0 \le CV \le 19$		≥ 16.5	
Bulk density	kg/m3		≥ 600			≥ 600	
Nitrogen content	% d.b	≤ 0.3	≤ 0.5	≤ 1.0	≤ 0.3	≤ 0.5	≤ 1.5
Sulphur content	% d.b	≤ 0	.03	≤ 0.04	≤ 0.05	≤ 0.2	≤ 0.4
Chlorine content	% d.b	≤ 0	.02	≤ 0.03	≤ 0.03	≤ 0.05	≤ 0.1
Ash melting behaviour	oC	≥ 1	200	≥ 1100	≥ 1200	≥ 1150	≥ 1000



Fig. 1. Typical pelleting process flow for wood and baled biomass.

plasticises and binds to create a polished appearance. This outer layer protects the pellet from adsorbing water vapour from ambient humidity [27]. In contrast, the ends are brittle and the main route for adsorption to occur. A number of studies have found that durable pellets were longer, though it is not clear whether this is a "cause or affect" observation. For example, increased tensile strength was observed with increasing pellet lengths across a range of feedstocks, but the shorter pellets had a higher MC and were less durable [28]. Another study showed a weak positive relationship between pellet length and durability and suggested that an indicator of durability could be the number of pellets per kg [13]. Another study found that an increase from 31.8 to 44.5 mm in the die depth significantly increased durability in wheat straw, corn stover and sorghum stalk pellets [29]. Changing the die thickness alters the diameter/length ratio, which discussed in Section 3.4.2.

## 3.2. Feedstock characteristics

## 3.2.1. Bulk density

The bulk density of the input material is an important factor in pelleting as the mills are fed by volume rather than weight [30]. Although few studies show a direct relationship between bulk density and durability, it can determine whether durable pellets are produced or not; therefore, this factor is mentioned briefly here. Low density is a particular concern with cereal residues and grasses. Loose straw has a density of 40 kg/m<sup>3</sup>, but this increases to 100–250 kg m<sup>3</sup> after grinding [31]. A suggested minimum bulk density is 200 kg/m<sup>3</sup> [32]; as steam-exploded cereal residues, with a bulk density of around 33–143 kg/m<sup>3</sup> did not form pellets in one study [33]. The bulk density of the resulting grind can be maximised by using the smallest possible hammermill size, and this is associated with a more durable pellet [31,34].

Pellet density is negatively correlated with the throughput of the plant, [33,35]. Also density tends to decrease with increasing MCs [36].

There is a very weak relationship between pellet density and durability [31,37], suggesting that high bulk densities do not imply proper compaction has occurred [38].

#### 3.2.2. Lignin

There is a strong positive relationship ( $r^2=0.68$ ) between pellet durability and lignin content [13]. Wood is typically composed of about 25% lignin, ranging between 15% and 40% across species [39], and can increase after biomass storage due to the decay of the readily available carbohydrate fraction [13]. The lignin content of *Miscanthus* and cereal straws is generally lower than wood (less than 20%), and therefore tend to produce less durable pellets [40–42]. Cereal straws and energy grasses also have higher ash contents (between 4% and 7%, [43]), which is undesirable.

Lignin is a complex phenolic polymer that provides mechanical strength to plant cell walls and protects from decay or invasion by pests and pathogens [39]. Both the quantity and composition is important for determining pellet durability. Lignin compositions vary between biomasses, and this affects the temperature at which it plasticises, also known as the glass transition temperature  $(T_g)$ . This mechanism enables the inter-diffusion of fibres and the formation of new bonds. The higher the Tg, the higher the temperature required to facilitate softening of the lignin. The higher the temperature above the Tg, the better, i.e. greater and easier, is the flow between fibres [44]. The T<sub>g</sub> of lignin and can range between 50 and above 100 °C [44], and the adsorption of water can reduce it [13]. Hardwood lignin tends to contain fewer phenolic hydroxyl groups and more methoxyl groups compared to softwood, and this has the effect of decreasing the T<sub>g</sub>. Therefore, when pelleting at the same temperature (100 °C) beech pellets form more solid bridges and show higher durability compared to Norway spruce [44].

#### 3.2.3. Extractives

Extractives include low molecular weight organic compounds, including fatty acids, waxes, terpenes and tannins [13,45]. Extractives have been shown to reduce pellet durability by lubricating the passage of material through pellet channels [46]. The decreased friction is associated with a lower energy requirement for pelleting [47], which could explain why softwood (with higher extractive content) is generally easier to pellet compared to hardwoods [46]. It is suggested that extractives also affect pellet durability by blocking binding sites for hydrogen bonding to occur between particles [36].

Nielsen et al., [26] found that Scots pine pellets, which contain more lipophilic extractives (5%), had much lower durability than beech (less than 1%). In another study, after 120 d of storage Scots pine produced more durable pellets than fresh material, this being more strongly attributed with a drop in extractive content ( $r^2$ =0.43) compared to lignin ( $r^2$ =0.11). A similar study found a markedly higher durability in pine that had been stored compared to fresh material [35]. Straw pellets were found to have poor durability due to high concentrations of wax at the surface of the material. This was believed to result in poor adhesion of particles [44], but is another example to how the presence of extractives can hinder durability.

Some studies show a different story. One found that fresh Scots pine showed a higher durability than stored material, with a positive relationship between extractive content and pellet durability, though they did observe decreasing power requirements, indicating lower friction, when pelleting material with higher quantities of extractives [47]. The authors suggest that extractives could have some role in pellet binding. A number of studies discovered that pellets produced from pure bark have excellent durability [13,45,48]. Bark usually contains higher levels of lignin than wood [48], but this depends on the age, region of the tree, and species [49], but it does contain far higher levels of extractives (Fig. 2). The effect of this can be quite complex, as one study [48] found that Scots pine pellets of blended assortments (5%, 10% and 20% bark) had a much lower durability than those made from



Fig. 2. Lignin and extractive content of wood and bark Scots Pine and Norway Spruce from different components of the tree (produced from data in [49]).

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100% pure wood or bark. In-homogenous shrinking of different sized particles found in the bark and wood blends may have caused this. Another study found that up to a 10% bark blend significantly reduced the durability of larch pellets, but showed slight improvements in wood types with a low lignin content [45]. Therefore, the negative effect of extractives could work against positive effects of lignin when bark is involved.

Bark is generally an undesired component in wood pellets as it contains considerably more nitrogen, sulphur and ash than wood, creating problems with emissions of nitrous oxides  $(NO_x)$ , sulphur dioxide  $(SO_2)$  and of slagging during combustion [48,50]. On a practical level, grinding bark can be hazardous as it creates a high proportion of fines, requiring particular health and safety procedures [48,51]. In response to a high demand for pellets in Europe, increasing numbers of pellet mills may utilise poor quality round wood; and it may be difficult to de-bark particularly small logs. Short rotation coppices are particularly difficult to debark and it is difficult to produce a good quality pellet from willow and poplar [52]. Therefore, the contribution of bark in pellets will potentially increase and could vary between each biomass supply chain.

## 3.2.4. Moisture content

Water has a crucial role in the pelletizing process and, along with lignin content, the MC of the feed is one of the most important parameters determining pellet durability [37]. As MC has both an antagonist and protagonist effects on durability, it must be optimised in the feed material. Higher MCs can reduce friction by lubricating the biomass [26]. Water is not compressible, however, limiting the final density of the pellet [53], and higher MCs increase the extent at which pellets 'relax' after formation, which can decrease durability [31]. On the other hand, moisture reduces the temperature at which lignin plasticises (Tg), which increases bonding between particles [46]. Above 20% MC it is suggested that steam pressure due to high temperatures reduces compression [47] or hydrogen bonds between wood polymers are substituted with bonds to water molecules, and the result is a weaker pellet [26,46].

The optimum MC ranges between feedstocks and studies, generally for pine it ranges between 6–13%, straw 8–15% and *Miscanthus* 20– 25% (Table 2). A number of studies on wood pellets show a positive correlation between MC and pellet durability. Between 8–15% MC, there is an increase in durability in Norway spruce and Scots pine [13]. Another study showed a positive correlation in durability ( $r^2$ =0.62) between a MC of 7–12% in scots pine [47]. The durability of larch pellets is shown to increase between a MC of 7–9%. Tulip wood pellets showed the highest durability at a moisture content of 13% [40]. The T<sub>g</sub> of hardwoods are generally higher than softwoods, so increasing MC can have a greater effect of improving durability [46]. Another study found that the durability of larch and tulip pellets increased steadily when the MC was increased from 9–17%, but again with large differences between the two wood types [45]. There were no differences

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Reported optimum moisture contents for pelleting different types of biomass.

Biomass Type	Product	Optimum M.C for pelleting (%)	Reference
Hardwood	Beech	8%	[26]
		10%	[44]
	Olive	5%	[53]
0 - 0 1	Gasta Dias	60/	[0/]
Sonwood	Scots Pine	6%	[26]
		8%	[35]
		11-13%	[44]
	Norway Spruce	10%	[44]
Cereal Residues	Wheat straw	8-10%	[57]
Cerear residues	Wheat Straw	15%	[44]
		10, 15%	[44]
		12–15%	[34]
Grasses	Miscanthus	20-25%	[32]

in the durability of wheat straw pellets between a MC of 9-14%, but after this point durability declined [29]. Across a range of biomass types including wood and straw, the optimum MC for pellet durability was between 6.5% and 10.8% [38]. In *Miscanthus* pelleting trials, it was found that higher quality pellets were produced at 20-25% MC [32]. This was also found in a recent study utilising a flat-die in the pellet mill, which found that a MC of 25% was optimal for pellet durability in *Miscanthus*, switchgrass and wheat straw pellets [54].

In contrast to above, a number of studies have reported a negative correlation between MC and pellet durability. In Nielsen et al., [26] pellets became progressively weaker from 5% to 14% MC, with the effect greater in beech ( $r^2=0.91$ ) than pine ( $r^2=0.76$ ). The authors suggest the surface layers of pine were 'contaminated' by a higher proportion of extractives than in beech, therefore the loss of hydrogen bonding capacity had a greater effect in beech. Another study found a gradually declining durability as the MC of olive pruning residues fell from 5% to 20% [53]. An optimum MC of 8% and a negative relationship was seen between durability and MC in Scots pine in another study [35]. Interestingly, one study [37] only observed a positive relationship between MC and durability after pine and spruce had been stored for 140 days. The authors relate the results to the lower extractive content of stored material, where the MC would then help binding. In biomass with higher extractive contents the additional water will only lubricate the material further, leading to a weaker pellet.

## 3.2.5. Summary: Feedstock characteristics

Some feedstock parameters have a greater effect on pellet durability than others. The lignin content is possibly the most important parameter, followed by moisture content, as these two factors directly interact to affect the temperature at which lignin softens. There are some conflicting results found in the effect of extractives on pellet durability: some studies suggest they lubricate the passage of material through the mill, whereas a few other studies suggest they have a role in binding. There is evidence that the effect of extractive content may be dependent on the particle size distribution and the lignin content. Changes in MC may also have positive or negative effects on durability, though it appears that there is some interaction with the extractive content.

## 3.3. Feedstock pre-treatment

## 3.3.1. Drying

Drying is sometimes necessary: as discussed in Section 3.2.4, ideally a woody feedstock should have a MC of 8-12% before entering the pellet mill [50]. Typically, freshly harvested wood has a MC of 50-55% [55], or around 30% after storage. Sawmill residues can range in MC depending on how long the logs have been seasoned. It is custom practice to saw timber prior to kiln drying, therefore sawdust could have a MC similar to that of freshly harvested wood, though smaller particles tend to dry out rapidly [56]. Energy grasses, such as *Miscanthus*, and cereal residues are usually harvested drier (up to 20%); in good weather conditions they may not require forced drying [57].

Obtaining the optimum MC of biomass prior to pelleting is vital to ensure pellet quality. Biomass dryers come in various types, and differ in heating techniques and temperature profiles. Varying methods are used in pelleting, such as hot air, desiccation, or vacuum drying, and hot-air driers are suited to industrial scales [50]. Wood chips dry at a slower rate than sawdust, but it is advantageous to dry prior to hammermilling as wetter feedstocks can cause sticking. Also, hammermilling wet material is reported to require more energy [58]. Sawdust is dried in a drum or flash drier, whereas chips are require lower temperatures and are best suited to flatbed driers [59]. If bark is used as a fuel source for drying then this could potentially contaminate the pellets [60].

Ståhl et al., [61] reviewed a number of convection driers commonly used to dry sawdust in Sweden, including those utilising flue gas, air or superheated steam as a drying medium. They found that driers with longer residence times caused greater losses of volatile organic compounds (VOC) in the biomass, which contribute to its calorific value. For example, sawdust dried in steam driers (240 °C) with a short retention time (2.5 min) lost 48–71% of their terpene content, whereas 80–83% was lost in a rotary drier with a longer retention time (110 h) and lower temperature (60–82 °C). Also, feedstocks with high initial MC lost more VOCs, mainly because they take longer to dry. Therefore, the drying technique may not affect pellet durability but the final LHV of the pellet.

#### 3.3.2. Hammermilling

Size reduction is a critical stage in pelleting. It affects a range of factors such as compaction, contact between particles, friction in the

#### Table 3

Screen sizes used in pellet studies and eventual pellet diameter (all in mm).

die and the flow rate of material [62]. A study examining olive tree pellets identified particle size as the third most important parameter in determining the hardness of pellets, the first two being a high temperature and optimised MC [53]. This was also found in Caribbean pine pellets (0.63 mm), with pressure and MC being the first and second most influential factors [63]. The particle size affects the total surface area, pore size and number of contact points for interparticle bonding required to produce durable pellets [33,64].

The extent of grinding required depends on the feedstock. In the case of sawdust hammermilling may not be necessary if particle sizes are generally less than 8 mm, so long that the material stream is screened for oversized pieces [56]. Good quality wood chips will comprise mainly (75%) of chip with a size range 3.15 and 16 mm, with no more than 12% less than 3.15 mm in size [65]. Straw and grasses are reduced to between 2.5 and 10 cm after shredding [66]. The final particle size is determined by a screen.

Size reduction is performed using a hammermill, which are rotary devices like wood chippers, but instead of being cut by blades the biomass is crushed by large metal hammers [67]. It is important that the final material is homogenous in terms of particle size and level of conditioning [25]. As mentioned above, the biomass is usually dried beforehand, but one study explored a wet comminution process involving *Miscanthus* and wood chips at 40% MC [42]. The method sheared rather than cut the biomass, and the highly fibrous product was found to produce a highly durable pellet. Likewise, a study on pine found that fibre orientation has a role in determining pellet properties [68]. Therefore, there is scope for exploring other methods of particle size reduction to produce stronger pellets.

Coarse particles sizes are ideal for combustion [34] and are classed as particles larger than 3.15 mm; fines are anything smaller. From a review of the literature, the majority of studies examining pellet formation use a maximum hammermill screen size of up to 4 mm (Table 3), though it ranges between 0.5 and 8 mm. Most suggest smaller screens sizes for optimum durability. A size limit of 5 mm limit is suggested [12], or a length of around 85% or less of the minimum thickness of the eventual pellet (between 5.1 and 10.2 mm [25]). Two references suggest ideal particle sizes of 0.5–0.7 mm [69] and 0.6– 0.8 mm [70], but are based on animal feed pellets. It is generally suggested that fine particles should not comprise more than 10–20% of the feed as it reduces the quality of the pellet and causes friction on the pellet die [22]. A high content of fine particles can also lead to blockages [71]. Small particles can dry out rapidly which can cause problems during pelleting [56].

Despite nearly every study mentioning the importance of particle size on pellet formation, few studies have explored the direct effect of particle size on durability. Overall, there is consensus that the finer the

Reference	Biomass type	Screen size (s) used	Optimum for durability?	Pellet diameter
[45]	Larch and Tulip wood pellets	1.41 < x < 3.17		7
[40]	Larch and Tulip wood pellets	< 1.41 and 1.41 < x < 3.17	< 1.41	7
[13]	Sawdust, bark and forest resides (pine and spruce)	3	_	6
[56]	Scots pine	1–8 mm	1 < x < 2	8
[48]	Scots pine	4	_	8
[42]	Scots pine and Miscanthus	2	_	6
[82]	Reed canary grass	6	_	8
[68]	Beech	0.5–2.8	2-2.8	10
[26]	Beech		_	11
[63]	Caribbean Pine	0.63-2	0.63-1	8
[53]	Olive tree residues	2-4	under 4	6
[28]	Olive tree residues	6 < x < 8	_	6
[29]	Corn stover, wheat straw, big bluestem	3.2-6.5	6.5	4
[52]	Wood, Miscanthus, cereal residues		3-6	6 to 8
[33]	Wheat, barley, oat and oilseed rape straw	0.8 to 6.4	0.8	6
[64]	Hay	2 to 6	4	6



Fig. 3. Durability of pellets produced from cereal residues with different hammermill screen sizes and with 25% addition of steam exploded biomass (produced from data in [33]).

grind, the more durable the pellet [22]. Pellets produced from fine particles show greater compression and abrasive strength [56]. Finer particles also allow for better particle flow, which has been demonstrated in spruce to give pellets with higher durability [62]. Cereal pellets showed a significant improvement in durability by reducing the screen size from 1.6 to 0.8 mm (Fig. 3, [31]). Another study on olive pruning residues found that particle sizes of 1 mm (vs 3 mm) were optimum for durability [53]. One study found this was true for larch, but not tulip wood, which is generally a poorer feedstock for pelleting [40]. There was an increase in pelleting friction as particle size decreased in beech, scots pine and wheat straw pellets [46], which is known to improve durability [26]. Some suggest that smaller particles rearrange and fill the voids of larger particles, improving the densification process [34], which is the case in particle board manufacture [56].

## 3.3.3. Binding additives

The use of binders has been explored in a number of studies (Table 4). They work as in a similar way as adhesive resins used in particle and fibre-board production [45]. They are added to improve combustion properties, improve durability, or to reduce wear on the pellet die [50,72], all of which can help to reduce the net GHG emissions from pelleting by reducing losses and decreasing the energy requirement of the process [14]. Current technical specifications require additives to be declared and must not comprise more than 2% of the total mass [19]. This limit seems to apply even if they are technically 'biomass' sources such as starch, flour or vegetable oils. Steam is the most commonly used binding additive in pelleting [17], though it is not usually considered to be an 'binder' *per se*, rather than a method of conditioning the feed.

A range of organic and inorganic binders have been explored in wood pellet production, however the eventual selection should be considered with costs and environmental impacts in mind [45]. When considering using a binder, one must consider the indirect environmental impacts that utilising a binder will have on pellet sustainability. For example, a recent study found that noticeable increases in the durability of wood pellets (Norway spruce and Scots pine) were achieved when using corn starch and molasses as additives, however the indirect GHG emissions from corn starch or molasses production were not compensated for by the improvements in durability [14]. That study found that even small uses of additives could not be justified through GHG mitigation alone.

A number of additives explored for wood pellet production have commonly been used in the animal feed pellet industry [22], such as sugary (sugar, molasses, cassava) and starchy additives (corn or potato flour), proteins, vegetable oils, or lignin or cellulose [19]. It is generally found that starch and water-soluble carbohydrates improve biomass pellet durability [15]. For example refined sugar and molasses, byproducts from sugar production, were shown to increase durability in wood pellets by 10–20% [15]. It is suggested these highly viscous components form strong bonds between particles that are similar to solid bridges.

Proteins can plasticise under heat, and have been shown to improve durability in beech pellets [71]. Proteins are generally higher value products, however and only have an important role in animal pellets [22]. Ahn et al., [45], found that rapeseed flour, coffee meal and lignin significantly improved the durability of larch and tulip tree wood pellets, which was attributed to the additives' high protein and starch content. There were differences between the two wood types, where a 2-10% binder had no real effect on larch but continuously improved tulip tree pellets. The authors suggest that tulip pellets had inherently lower durability, due to a lower content of lignin. Bark and pine cones had a negative effect on durability in larch and a small benefit in tulip tree, maybe due to the higher extractive content of these materials. A study examining blended Miscanthus and sawdust pellets found that a high durability (97.5%) was only achieved with a 2% potato starch additive. Again, this was due to the lower lignin content of Miscanthus [42].

A recent study examining a 2.2% and 5.8% blend of waste vegetable oil with wood pellets found that the oil increased the energy value of the produced pellets, but it also lubricated the material, reduced the pelleting energy requirements, and thus decreased the resulting durability [73]. It is possible that oil-based coating agents can be applied after pellet formation, which can provide effective protection from external moisture and increase the LHV [74].

Non-food based additives have also been explored in the literature. Lignin, already identified as a key determinant of pellet durability, can be added to feedstocks where the relative content is low. For example, a 2% addition of kraft lignin and lignosulphate, both by-products of the paper industry, have been shown to increase pellet durability in Scots pine by 0.8% [75] and Norway Spruce by 1.4% [76]. In another study, wheat straw pellets made with a 5% glycerol, 2% bentonite (aluminium phyllosilicate) and 2% lignin addition showed improved strength [77]. Glycerol has also been explored in wheat, barley, oat and canola straw, which did not show a marked increase durability, but did show a lower ash content and increased higher heating value [78]. In another study, increasing concentrations of algae significantly increased the compressive strength of *Miscanthus* pellets, however adding more than 20% caused a significant decline in LHV [79].

#### 3.3.4. Steam explosion

Steam explosion uses high pressure steam to 'activate' lignin for binding [80]. It has been explored as a pre-treatment process in lignocellulosic bioethanol production, as it increases accessibility of cellulose to hydrolysis, but some studies have explored the use of steam explosion to improve the formation of pellets from wood [27] and cereal residues [33]. In willow, steam pre-treatment produced pellets with good strength and reduced ash content, improving the combustion properties [27]. A thesis studying the effect of steam explosion on Douglas fir found improved durability with treated pellets [81]. Tests on cereal residues found that steam pre-treatment durable pellet increased the bulk density of the biomass to such an extent that feeding problems occurred in the pellet mill and reduced durability (Fig. 3), despite smaller scale tests showing improved pellet durability after steam explosion [33].

## 3.3.5. Summary: Feedstock pre-treatment options

This section describes how biomass is prepared before entering the pellet mill. The main stage affecting pellet durability is size reduction. Although a range of particle sizes are reported in literature, it is generally found that smaller particles produce more durable pellets as they increase friction in the mill and can occupy voids more effectively than coarser particles. Obtaining the optimum MC of biomass by drying is vital to ensure pellet quality, however this is discussed in the previous section. Otherwise the method of drying mainly affects potential losses of VOCs, which affects the eventual LHV of the pellet. Binders can improve durability in pellets, particularly those with low lignin contents. Oily or fatty binders should be avoided as these lubricate the material and make a weaker pellet. Steam conditioning can help activate lignin but care must be taken not to increase the bulk density of the biomass so that it negatively affects durability.

## 3.4. Pelleting conditions

## 3.4.1. Temperature

Preheating the feed material activates the binders present and promotes the deformation of thermoplastic particles that are necessary for pellet formation [22]. This section specifically refers to the temperature of the material entering the pellet press. Wood pellets require temperatures of 110–130 °C for to binding occur [26]. In wheat straw, the  $T_g$  is between 53 and 63 °C [41], though it is still recommended that a pressing temperature of 100 °C is used as lower temperatures fail to overcome the effect of a high level of extractives in straw. An optimum temperature of 105 °C is suggested for *Miscanthus* [32]. Heating can be provided from the drying equipment, or from supplementing the flow with steam, (Table 4), or providing heat indirectly through conduction based heating systems [22].

It is generally found that pelleting temperatures over the specific biomass'  $T_g$  will aid durability [22]. Lee et al., [40] found that increasing temperatures positively affected the durability of larch. Temperature increased durability of beech and scots pine [26], and in beech, Norway Spruce and straw [44]. Carone et al., [53] found that temperature was most important variable influencing pellet mechanical properties in olive pruning residues. Interestingly, the opposite effect was seen in reed canary grass pellets, where lower die temperatures (30–45 °C vs. 65 °C), gave optimal pellet durability after continuous pelleting [82]. The authors found that although high temperatures increased the flow rate of material through the mill, this caused problems with irregularities in the feed, but this is a general problem with pelleting straws and grasses [83].

## 3.4.2. Pelleting pressure

The physical forces that build up in the pellet die are crucial for understanding and optimising the pelleting process [12]. This section describes the pressure required to overcome the force of friction from the material passing through the mill [26]. The pressure applied between the rollers and the die can be affected by a number of factors including the motor power, the rolling speed, the bulk density of the feed, and the dimensions and fabric of the pellet channel [17]. It can also change during the life of the die, for example a new pellet mill will run at 4.5 t/h but half worn it may need to run at 3.5 t/h to maintain enough pressure to achieve the required pellet quality [24]. Generally, wear occurs at a faster rate when pelleting straw compared to wood [25]. Oily/fatty binders used to help improve the speed of passage of material through the mill [50] have an adverse effect on pellet durability due to the reduced pressure applied in the mill [73]. Two studies suggest an interaction between pressure and temperature and MC [26,46], finding that heat and moisture can smoothen the flow of material through the die, therefore would need to be optimised in some feedstocks to ensure durability is achieved.

Pellets are produced at pressures between 115 and 300 MPa (Table 4), and generally, higher pressures give more durable pellets [26]. Two studies using very low pressures (1.5 MPa) produced poorer pellets compared to standard pellets [40,45]. Higher pressures increase durability in cereal residues [34] and reduce pellet relaxation after formation [31]. A study on olive pruning residues found no difference in durability in pellets produced between 70 and 175 MPa, though interactions between pressure and other factors suggested 170–180 MPa was optimal [53]. Another study suggested that only marginal improvements in durability could be achieved in beech and Scots pine above 250 MPa [46]. Beyond a certain pressure the compaction of the pellet is limited to the relative density of the material [84].

The length/diameter ratio of the of the pellet channel is a good metric for the degree of compression experienced by the material during pelleting [80]. Die diameters tend to be constrained by the specification limits for pellets (6–10 mm). The lengths of the pellet dies do not particularly affect the length of the pellet and range between 20 and 180 mm (Table 4). It is found that higher L/D ratios exponentially increase the pressures in the mill [46,71] and are shown to improve pellet durability [29,32]. This is particularly important in feedstocks with low lignin content, such as cereal residues [25].

## 3.4.3. Cooling

When pellets leave the pellet mill they have a temperature of 70– 90 °C [52]. As the binding mechanisms in pellets relies on the melting and re-solidifying of lignin, pellets do not gain their true strength until they have cooled [17]. This is done by blowing cooled (0–25 °C) air over newly formed pellets until they reach within 5 °C of ambient temperatures. Effective cooling is necessary as cracks caused by the temperature gradient between the outer and inner layers can lead to problems with fine production and breakage [13].

## 3.4.4. Summary: Pelleting conditions

In general, pelleting temperatures of between 100 and 130 °C, though this is dependent on the specific  $T_g$  of the lignin in the biomass. High pressing pressures between 115 and 300 MPa are required to produce durable biomass pellets. A couple of studies suggest that increasing the temperature and MC can reduce pressure through the mill, highlighting the need to optimise these three parameters for different feedstocks. Effective cooling is necessary to ensure that the pellets solidify without causing breakage.

## 3.5. Post-pelleting events

The main challenge of producing pellets is making them sufficiently strong enough to endure mechanical wear caused by handling [17]. Pellets are handled and dropped between eight and ten times between being produced and reaching their final destination [85,86]. Breakage occurs either from cracks developing within the pellets or from abrasion [87]. In supply chains pellets are either conveyed, dropped or blown into trucks, bunkers or vehicles. It is believed that dropping wood pellets causes breakages and increases fines, yet there is no standard protocol for drop tests for pellets [86].

A study tested pellets with a 97% durability in various drop tests. They found that a single 22.8 m drop to a concrete base led to a mass

<b>Table 4</b> Overview of	parameters used in pelle	ting studies.	Those highligh	tted in bold ar	e the test para	ameters.					
Reference	Feedstocks	Pressure (MPa)	Channel Diameter (mm)	Channel length (mm)	Temp. (C)	Moisture content (% w.b)	Binders	Drying Method	Steam used?	Durability of Pellets	Main conclusions
[45]	Larch Tulipwood	1.5	~	120	180	13–19%	Rapeseed flour Coffee meal Bark Pine cones Lignin powder	1	None	95.1–97.5*%	Larch improved with rapeseed flour, coffee meal and lignin powder. Tulip wood 95.1, improved with all binders. Higher moisture contents gave higher durability
[40]	Larch Tulipwood	1.5	~	120	120-180	9 to 13	None	I	I	Larch – 97.9 –99.6 Tulip wood- 94.9 – 96.2	A longer pelleting time and increasing temperature increases durability. Smaller particle sizes increased durability
[35]	Norway spruce Fresh and stored	I	I	65	I	8.2-11.7%	I	I		98.1–99.3	Lower moisture content and stored material gave highest durability
[56]	scots pine Scots pine at different particle sizes	I	ω	55	73-103	I	1	I	I	'Small particles' 0.8– 1.5% (corresponds to durability of 98.5– 99.2%)	Finer particles gave higher durability.
[47]	Scots pine (fresh and stored)	1	∞	1	1	7-13%	I	75 C warm air- batch drier 450 C warm air-batch	Without and with steam	7% M.C- 92% 12% M.C- 96%	Moisture content increased durability (optimum 10–11%). Higher extractive content increased durability. Steam had no effect. Fresh material had higher durability than stored.
[26]	Scots Pine Beech	300	œ	I	60-160	11.3	1	arter Saturated salts	None	(measured pellet strength)	Temperature and MC reduced pressure required to press material into channel. Temperature and MC increased pellet
[68]	Beech Aspen Scots Pine	115	10	I	120	12	I	Desiccator at 75 C	I	(measured compression energy)	suctuation (Examined fibre orientation) Found that transverse fibre orientation improved production capacity and reduced energy consumition
[28]	Olive tree Almond tree Black poplar	1	6	1	200	lower than 15%	None	Rotary drier (250 C)	I	~ 85% of sample passed CEN standards	Longer particle sizes lead to stronger pellets. Moisture content affects particle density which affects durability.
[44]	Houn Oak Beech Norway Spruce Wheat straw	200	œ	16	20 to 180	10	I	Vacuum dried at 40 C for 2 days	I	(measured force of break)	Pellets produced at 100 C have higher mechanical strength than 20 C. At 100 °C beech produces better pellets, Tg of Norway spruce requires higher temperature and straw is covered with waxy additive layer.
[42]	Sawmill residues	I	9	20	100-115	15%	Miscanthus – (< 2%) Potato starch		none	96.5 - 99.3%	Motsture content timit is 1.5%. Starch binder required to achieve durability of over 97.5%. Miscanthus had little effect.
[33]	Wheat straw, Barley straw Oat straw OSR straw	31.6 to 138.9	6.25	135.3	95	10%	(0.4)	I	180 C (900 kPa for 4 min)	Non treated 89–95% Steam exploded (25%) 82–95%	Decreasing hammermill sizes increased durability. Lower pressures lead to higher relaxation and lower durability. Steam exchosion did not innerver ballet durability.
[34]	Wheat straw Barley straw Corn Stover Switchgrass	40-160	I	I	100	12 to 15	I	Not required	I	(measured compression and relaxation)	Pelletaensity increased with increasing compressive pressure. Particle size and moisture content both influenced mechanical properties of pellets. (continued on next page)

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Reference	Feedstocks	Pressure (MPa)	Channel Diameter (mm)	Channel length (mm)	Temp. (C)	Moisture content (% w.b)	Binders	Drying Method	Steam used?	Durability of Pellets	Main conclusions
[32]	Miscanthus	I	24-30	Q	105	10 to 30	None	Not required	None	35.9–97.4	Optimum conditions: bulk density 240– 300 kg/m3, die ratio of 4.5:1 to 5:1 and 20– 25% M.C
[53]	Olive tree pruning residues	71-176	9	12	60-150	2-20%	I	Oven dried (65 C)	I	(measured modulus of elasticity)	High temperatures, low moisture content and small particle sizes increase pellet durability.
[29]	Wheat straw Big Bluestem Corn Stover Sorghum stalk	I	4 and 6.4	31.8 and 44.5	74-82	10%	None	SII	None	(WS) 95.8–98.3 (BB) 95.7–97.6 (CS) 96.4–98.2 (SS) 85.7–93.5	Moisture content above 14% decreased durability. Larger hammermill screen size increased durability Longer pellets were more durable.

Fable 4 (continued)

loss of 1% as fines, increasing to 10% after five repeated drops; showing how increased handling frequency can accelerate the rate of breakage [86]. The authors discovered a linear relationship between pellet breakage and the height at which pellets were dropped. Therefore, durable pellets can be somewhat preserved by careful handling during transport and storage. Shipping ports that are adapted to handling large quantities of wood pellets have established handling protocols to reduce damage to pellets, such as specialised grabbers, low-speed conveyors, low drop heights and no handling during wet weather [88]. When delivering pellets to a storage silo, the process of blowing pellets into the hold can damage them, though blowing at an angle of 15–20° onto an impact protective mat will reduce this [89].

It is very important to ensure that the storage facilities are watertight, as exposure to moisture can cause pellets to swell and disintegrate [90]. Storage silos need to provide protection from rain, condensation or ground water [17,89]. Some self-heating can occur in pellets due to chemical oxidation of the biomass, or from physical forces of condensation and adsorption [91,92]. This can lead to the condensation of water causing pockets of damp, usually in a centralised 'chimney' [93]. As this is a slow process, it may only become problematic at large scales, and can be suppressed by flooding the storage areas with nitrogen gas [88]. It is possible to protect pellets by immersing them in vegetable or mineral oils for up to 10 s. Pellets treated in such way have the ability to withstand exposure to moisture levels that would rapidly disintegrate untreated pellets [74]. The oil coatings also increased the LHV of the pellets by 5.7% but the application levels will most likely exceed the 0.2% coating limits specified by the EN-14961-2 standards, and the environmental tradeoff would need to be determined.

## 4. Summary of observations

Examining and identifying the factors affecting the durability of pellets is difficult because it is affected by many factors such as MC, particle size, addition of binders and pelleting conditions [94]. Although there is a standard test for 'durability', some studies refer to compressive or abrasive strength, elasticity or water resistance, where one must assume that pellets that show high strength or resistance are highly durable. Generally, durable pellets are formed when there is successful development of covalent, non-covalent bonds and Van der Waals forces between adjacent polymer chains of the biomass. Although the pelleting conditions depend on specific biomass characteristics, a number of factors can be summarised that increase both the binding mechanisms and friction of the material in the pellet channel (Fig. 4). While some factors improve binding and some increase friction, some affect both binding and friction. Improving binding mechanisms are of higher importance for pellet quality, high pressure and friction does not alone guarantee the production of durable pellets [22]. Across studies there is a general consensus that the higher the pressure, the greater the contact is between particles, and the denser and more durable the pellet [33,53]. Increasing friction in the die can increase the energy demand and the wear of equipment, which can have implications on the energy balance of the system. Increasing the temperature can reduce wear on the die while aiding the plasticising of lignin [26], but will increase the energy demand of the process. Overall, durable pellets are correlated with high energy consumption [31], suggesting this is an unavoidable trade off if durability is to be prioritised. This affect may be compensated for by using binders to increase adhesion while reducing the energy demands for pelleting, however when using food-based additives there is evidence that this does not improve the overall GHG balance of the pellets, mainly due to the indirect GHG emissions from manufacture of the binders [14].

It is possible to produce durable pellets from wood, cereal residues and energy grasses. Woody feedstocks showing excellent durability include Scots pine, Norway spruce and beech. Softwood is believed to



Fig. 4. Summary of factors affecting pellet binding mechanisms and friction in the pellet channel.

produce higher quality pellets than hardwood as it has a higher lignin content, though good quality hardwood pellets can be produced when binders supplying additional lignin are used. Wood is far easier to pellet than cereal residues and energy grasses, as these have a lower lignin content, higher extractive content and the waxy surface layer on cereal residues can hinder pellet formation. Despite this, straw has been described as a 'more attractive' feedstock for pelleting as it is usually harvested at a lower MC than wood, and if the seasonal conditions are optimum it may not require artificial drying before it can enter the pelleting process, reducing the energy balance from producing them [57]. A disadvantage of using cereal residues is that they tend to contain higher nitrogen, sulphur, chlorine and potassium and thus ash contents, which can lead to slagging and corrosion of boilers [25]. These contaminants are due to the agronomic inputs applied to guarantee the yields of their cereal grain counterparts, so could affect energy grasses if artificial fertilisers are used during their growth [95]. Assessing the chemical composition of pellets from different sources of biomass is out of scope of this study. Bark may produce a durable pellet; however, it also contains undesirable concentrations of nitrogen, sulphur and ash. Short rotation coppices are particularly difficult to debark and it is difficult to produce a good quality pellet from willow and poplar.

## 5. Conclusion

A high lignin content and optimum MC coupled with high pelleting temperature tends to improve biomass pellet durability. Conversely, coarser particle sizes, high extractive contents and high MCs reduce durability by reducing friction and disrupting binding. Some biomass qualities may change during storage, therefore it is suggested that an optimal strategy would be to separate stored and fresh material. There is some evidence that starch or lignin-based binders can improve durability, particularly in feedstocks with lower lignin contents. Adding fat or oil-based binders might increase the LHV of the pellet and reduce the energy consumption of compression, but the reduced friction means that the pellets are more elastic and less durable. Wood is generally easier to pellet than cereal residues and energy grasses, which has been attributed to a high lignin content and lower extractive content.

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