Swedish University of Agricultural Sciences
The Faculty of Forest Sciences
Unit of Biomass Technology and Chemistry

Effects of storage time, die channel length and moisture content on pellet quality of lodgepole pine (*Pinus contorta* Dougl. var. *latifolia*)

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Photo: Olof Högqvist

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Effects of storage time, die channel length and moisture content on pellet quality of lodgepole pine (*Pinus contorta* Dougl. var. *latifolia*)

Effekten av lagringstid, matrislängd och fukthalt på pelletskvalitet av contortatall (*Pinus contorta* Dougl. var. latifolia)

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Abstract

The use of renewable resources is rapidly increasing in Sweden, especially the use of fuel pellets. Through pelletizing the energy content per volume unit is greatly increased and the pellets are more homogeneous with regard to moisture content, particle size and density compared to unrefined fuels like forest residues and chips. For further development of the Swedish pellet industry, the use of different assortments from lodgepole pine (*Pinus contorta* Dougl. var. *latifolia*) is of particular interest, mainly because of its high volume production. Whole tree and stemwood assortments can be chipped, milled and processed to fuel pellets but many properties have to be fulfilled to achieve a high quality pellet. Pellet quality is a result of chosen raw material with its properties and chosen process settings. The aim for this study was to investigate how the factors raw material moisture content (%), die channel length (mm) and storage time (days) effects the responses bulk density (kg/m^3), mechanical durability (%), fines (%) and energy consumption (A) for the assortments stemwood and whole tree of lodgepole pine. Bark was included in both assortments and the whole tree assortment also contained branches, tops and needles.

A three-dimensional Central Composite Face (CCF) design was setup to study the effects of the three factors on the four responses. The effects of storage time on extractive contents, ash contents and ash melting temperatures were also analyzed. The results showed that the stemwood assortment could be predicted with high accuracy for all responses but the whole tree model gave low prediction accuracy. The result for the stemwood assortment showed that bulk density increased with longer die channel length and higher moisture content within the intervals of 45-55 mm and 9-13 %, respectively. Durability and energy consumption increased while fines decreased with increased moisture content. A storage time of around 200 days gave the optimum values for bulk density, durability and fines. A significant decrease of extractives was observed between storage time 242 and 325 days. Mean extractive content were 2.64 % in stemwood and 3.89 % in whole tree assortment. Mean ash content were in stemwood 0.42 % and 0.61 % in whole tree assortment. These ash contents pass the requirments stated in the European standard for the highest quality class of fuel pellets. Ash melting temperatures were around 1500 °C for both stemwood and whole tree assortments.

The conclusion is that moisture content was the most significant factor explaining the durability and the proportion of fines and the relationships were positive and negative, respectively. The relationship between storage time and all responses were positive (up to 200 days) with the exception of energy consumption. Die channel length was one of the most significant factors that affected the bulk density and energy consumption and the correlation was positive. The overall conclusion is that stemwood (including bark) of lodge pole pine within the found production window can be used as feedstock to produce fuel pellets of high quality.

Sammanfattning

Användningen av förnyelsebara resurser ökar snabbt i Sverige, särskilt användningen av bränslepellets. Genom pelletering ökas energiinnehållet per volym enhet och pelletterna är mer homogen vad gäller fukthalt, partikelstorlek och densitet jämfört med oförädlade bränslen såsom GROT (grenar och toppar) och flis. För fortsatt utveckling av den svenska pellets sektorn kan användningen av olika sortiment av contortatall (*Pinus contorta* Dougl. var. *latifolia*) vara av särskilt intresse mest på grund av trädslagets höga volymproduktion. Helträd och stamvedssortiment kan flisas, malas och bearbetas till bränslepellets, men flera egenskaper måste uppfyllas för att uppnå en hög pelletskvalitet. Pelletskvalitet är ett resultat av den valda råvaran med dess egenskaper och valda processinställningar. Syftet med denna studie var att undersöka hur faktorerna råvarafukthalt (%), matrislängd (mm) och lagringstid (dagar) påverkar responserna bulkdensitet (kg/m^3), mekaniskt hållbarhet (%), finfraktion (%) och energiförbrukning (A) för sortimenten stamved och helträd av contortatall. Bark ingick i båda sortimenten och helträd innehöll även grenar, toppar och barr.

En tredimensionell Central Composite Face (CCF) design skapades för att studera effekterna av de tre faktorerna på de fyra responserna. Eventuella effekter av lagringstiden på halten av extraktivämnen, askhalt och askans smältförlopp analyserades även. Resultaten visade att stamvedssortimentet kunde predikteras med hög säkerhet för alla responser men att helträd gav låg prediktionssäkerhet. Resultatet för stamvedssortimentet visade att bulk densiteten ökade med längre matrislängd och högre fukthalt inom intervallen 45-55 mm och 9-13%, respektive. Hållbarheten och energiförbrukningen ökade och andelen finfraktion minskade med ökad fukthalt. En lagringstid på cirka 200 dagar gav de högsta värdena för bulk densitet, hållbarhet och andel finfraktioner. En signifikant minskning av halten extraktivämnen observerades mellan lagringstid 242 och 325 dagar. Den genomsnittliga extraktiv halten var 2,6% i stamved och 3,9% i helträdssortimentet. Den genomsnittliga askhalten var i stamved 0,42% och 0,61% i helträdssortimentet vilket innebär att askhalten passerar de krav som anges i den europeiska standarden. Asksmälttempraturen var cirka 1500 °C för både stamved och helträdssortimentet.

Slutsatsen är att fukthalten var den mest signifikanta faktorn som förklarade hållbarheten och andelen finfraktioner och sambanden var positiva och negativa, respektive. Sambandet mellan lagringstid och alla responser var positivt (upp till 200 dagar) med undantag av energiförbrukningen. Matrislängden påverkade bulk densiteten och energiförbrukningen och sambanden var positiva. Försöken visar att speciellt stamved innehållande bark av contortatall kan användas för produktion av bränslepellets av hög kvalitet inom det funna produktionsfönstret.

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Olof Högqvist

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1 Introduction

The bioenergy market in Sweden is rapidly growing and the use of renewable resources currently contributes to 47 % of the energy demand (Energimyndigheten, 2011) including 22 % of biofuels, peat, waste etc. (Skogsstatistisk årsbok, 2011). Within the bioenergy sector the pellet market are booming. From 1997 to 2010 the amount of fuel pellets delivered to the Swedish market has increased from 2.4 to 10.9 TWh (Energimyndigheten, 2011).

Converting biomass to pellets greatly increases the energy density compared to unrefined fuels (Obernberger & Thek, 2010). Pellets have lower moisture content and are more homogeneous with regard to both moisture content and particle size (Obernberger & Thek, 2010). Pellets can be used in many energy conversion processes from small households to industrial use (Obernberger & Thek, 2010). The regular fuel quality makes it possible to convert oil and gas burners in small households to almost entirely wholly automated systems (Obernberger & Thek, 2010). These properties make altogether fuel pellets easy to handle, transport and burn (Filbakk et al., 2011a).

The main raw materials for the pellets industry are by-products (sawdust and cutter shavings) from the sawmills (Helby et al., 2004). But a competition for these products has begun, and the produced by-products are not enough to fulfill the demands for the pellet industry (Selkimäki et al., 2010). Shortages are predicted to be more common in the future and new raw materials has to be found to facilitate further development in the pellet sector (Selkimäki et al., 2010).

Possible assortments to meet higher demands could be low quality tree stems or residues from the forest (Sikkema et al., 2011). One particular biomaterial of interest in Sweden is the lodgepole pine (*Pinus contorta* Dougl. var. *latifolia*). In Sweden lodgepole pine has on average 36 % higher volume production than the domestic Scots pine (*Pinus sylvestris* L.) independent of site index (Elfving et al., 2001). Large areas of lodgepole pine were planted during the 1970-80's in Sweden (Skogsstyrelsen, 2009) and are now in the first thinning stage. In 2010, there were around 640 000 ha of lodgepole pine plantations in Sweden, which represents approximately 0.4 % of the total timber storage (Skogsstatistisk årsbok, 2011).

Whole tree and stemwood assortments can be milled and processed to fuel pellets but several properties have to be fulfilled to achieve high quality fuel pellets. Whole tree assortments have not been pelletized in bigger scale but combinations of bark ratios have been done (Filbakk, 2011b; Bradfield & Levi, 1984; Lestander, et al., 2012). Whole tree harvest (forest residues) contributes to 15 % of Sweden's total energy use (Egnell, 2011) and stemwood and forest residues are generally harvested separately in Sweden.

1.1 Pellet quality parameters

In Europe, pellet quality standards are stated by the European Committee for Standardization (CEN). The standards are either normative or informative. For normative standards only quality classes are defined. In informative standards information must be stated and given to the customer. The standards are divided in different quality classes. The specifications (normative and informative) of the highest pellet class for non-industrial use class A1, are shown in Table 1.

Table 1. Specifications, specification type and desired values for the European Standard EN 14961-1: 2010 and stated values for the highest quality class A1 EN 14961-2: 2011.

Specification	Specification type	Value class A1[a]	Desired value
Orgin and source	Normative	Specified	Specified
Pellet dimensions	Normative	D 6 or 8 and L ≥ 3.15 ≤ 40 mm[b]	Low
Moisture content	Normative	≤ 10 %	Low
Ash content	Normative	≤ 0.7 %	Low
Mechanical durability	Normative	≥ 97.5 %	High
Amount of fines	Normative	≤ 1 %	Low
Amount of additives	Normative	≤ 2 %	Low
Net caloric value	Informative	≥ 16.5 ≤ 19.5 MJ/kg or Kwh/kg	High
Bulk density	Informative	≥ 600 kg/m^3	High
Chemical content	Informative	Specified	Low
Ash melting point[a]	Informative	Specified	Specified

a) The higest quality class for non-industrial use EN 14961-2: 2011.
b) D = Diameter; L = Length (mm).

1.1.1 Ash Content

Ash contents in wood are depending on soil type, tree age, tree section (needles, twigs and bark) and season of harvest (Petersson & Nordfjell, 2007). A tree absorbs minerals from the soil and allocates it mainly to its needles or leaves, small diameter twigs and bark (Petersson & Nordfjell, 2007). In softwood ash contents are around 5 % in needles, 3 % in bark and 0.5 % in stemwood (Petersson & Nordfjell, 2007). So therefore assortments with high proportion of bark and needles will result in high ash contents. Differences in proportions of these groups together with contamination of mineral soil will affect the final ash content (Petersson & Nordfjell, 2007). The European standard EN 14961-2: 2011 states that the ash content should be ≤0.7 % of dry weight for the highest quality class pellets, A1 (Table 1). Koch (1996) has reported ash contents from 0.23-0.28 % in lodgepole pine stemwood and Lindström et al. (2010) has reported ash contents of 1.1 % in Scots pine (*Pinus sylvestris* L.) whole tree assortment.

1.1.2 Ash melting temperatures

High ash contents can create problems in burners due to ash formation elements that lead to severe slagging (Lindström et al., 2010). Two types of slagging tendencies are common, glass and salt formations (Ericson, 2006). Glass formation is formed when ash oxides (K_2O and CaO) react with silica (SiO_2) which is melting in low temperatures (Ericson, 2006). Potassium, calcium and sodium are the reactive components in salt formations (Ericson, 2006). In both these cases the formations are creating sticky areas where more ash and dust emissions are get stuck (Ericson, 2006) and are especially a problem in fluidized beds (Werkelin et al., 2005). These problems are in general related to lower combustion efficiency and higher emissions of particles (Ericson, 2006). The ash melting temperature is an indication of how the fuel ashes will react in the burner. A low melting point can lead to severe slagging so a high melting point above 1300 °C is preferred (Löfgren, 2004).

1.1.3 Mechanical durability

Mechanical durability is one of the most important parameters in pellet production and is defined in European Standard EN 14 588 as “ability of densified biofuel units (e.g. briquettes and pellets) to remain intact during loading, unloading, feeding and transport” (Obernberger & Thek, 2010, p. 51). The standard test for determination of mechanical durability is performed by tumbling the pellets and then calculate the percentage of remained pellets after removing the fines. Mechanical durability should be ≥97.5 % for the highest quality class A1, see Table 1 (EN 14961: 2010).

1.1.4 Amount of fines

A low durability results in high amounts of fines which can lead to problems during storage, feeding and combustion processes (Filbakk et al., 2011a; Obernberger & Thek, 2010). During storage and handling high amounts of fines can create dust explosions (Obernberger & Thek, 2010) and therefore affect the user comfort (Filbakk et al., 2011a). In the feeding process high amounts of fines can block the feeding screws by forming stable structures across openings, so called bridging (Jensen et al., 2004) which causes stop in the production line. Furthermore particulate emissions and fuel residues increases when high amounts of fines are present in the combustion processes (Filbakk et al., 2011a; Obernberger & Thek, 2010). For the highest quality class A1 in EN 14961-2: 2011 the amounts of fines should be ≤1 %, see Table 1.

1.1.5 Bulk density

Pellet bulk density is weight divided by the bulk volume (kg/m^3). A high bulk density results in high energy content per bulk volume which leads to lower transport and storage costs (Obernberger & Thek, 2010). In the European Standard EN 14961: 2010 it is specified that pellets should have a bulk density ≥600 kg/m^3 for the highest quality class A1 (Table 1), but the pellet producers often aiming for 650 kg/m^3 (Obernberger & Thek, 2010).

2 Bounding properties

The bounding properties are important for pellet durability (Mani et al., 2003). The fundamental knowledge for how particles in pellets bound together is still relative unknown (Mani et al., 2003). Rumpf (1962) was the first to describe possible bounding mechanisms in granules and agglomerates. Rumpf (1962) proposed the following five mechanisms.

1. Attraction forces between solid particles e.g. hydrogen boundings and Van der Waals forces.
2. Interfacial forces and capillary pressure in movable liquid surfaces.
3. Adhesion and cohesion forces.
4. Solid bridges.
5. Mechanical interlocking between particles.

Attraction forces between solid particles are short range forces that are active only when particles are close together and the attraction decreases rapidly when the distance is increasing (Mani et al., 2003). The forces can be valence, Van der Vaals and magnetic. The strength depends on particle size, surface charge, crystal structure, proximity to other particles, the amounts of binders and other physic-chemical properties (Mani et al., 2003).

Interfacial forces and capillary pressure in movable liquid surfaces are results of surface tension and capillary forces between the liquid and the particles. These forces create strong bonds between particles but disappear when the liquid evaporates (Mani et al., 2003).

Cohesion is the attraction of molecules of the same substance to stay together while adhesion is the attraction of molecules from different substances to stay together.

Solid bridges are formed under high pressure and temperature by crystallization of dissolved substances, hardening of binders, melting & sintering and chemical reactions (Mani et al., 2003).

Mechanical interlocking is how particles mechanically fit together and are suggested to have a minor contribution to pellet strength (Mani et al., 2003).

3 Process settings

The quality of pellets can be depending on process settings like die channel length (Holm et al., 2006; Holm et al., 2007) and raw material moisture content (Grover & Mishra, 1996; Obernberger & Thek, 2010; Kaliyan & Morey, 2009; Filbakk et al., 2011a; Mani et al., 2003; Nielsen et al., 2009; Samuelsson et al., 2009; Rhen et al., 2005; Lehtikangas, 2001).

3.1 Raw material moisture content

When manufacturing densified products (e.g. pellets) the material is compressed under high pressure (Grover & Mishra, 1996). In this compaction stage water has a crucial role (Grover & Mishra, 1996). In pellet production the moisture content of the raw material are controlled by either drying the material or adding steam before pelletizing (Obernberger & Thek, 2010). Water can acts as both a binding agent that affect the durability properties and as a lubricant that lower the energy consumption (Kaliyan & Morey, 2009).

A review article including all kinds of biomaterials concluded that strength and durability increasing with increasing moisture content until an optimum are reached (Kaliyan & Morey, 2009). The optimum value varies for different raw materials and process settings (Filbakk et al., 2011a). In woody materials moisture contents from 8-12 % are often desirable to achieve high quality pellets (Obernberger & Thek, 2004).

Van der Waals forces and hydrogen bounds are suggested to act like bounding mechanisms when particles are compressed (Grover & Mishra, 1996; Mani et al., 2003). Water are acting like a bridge between the wood particles and therefore increasing the area for bounding opportunities between these particles (Kaliyan & Morey, 2009; Grover & Mishra, 1996; Mani et al., 2003). At the same time water can sorb to hydrogen bounding sites and therefore occupying the site for particle-to-particle bounding so high amounts of water can decrease the bounding opportunities (Nielsen et al., 2009).

In wood pellets a negative correlation have been reported between moisture content and pellet strength (Nielsen et al., 2009), compression strength (Rhen et al., 2005), durability (Samuelsson et al., 2009), bulk density (Samuelsson et al., 2009; Filbakk et al., 2011a) and energy consumption (Samuelsson et al., 2009; Filbakk et al., 2011a; Nielsen et al., 2009). And a positive correlation to fines (Samuelsson et al., 2009) and durability (Lehtikangas, 2001).

3.2 Die channel length

The dominant process technology in production of fuel pellets is to feed the raw material vertical through a ring-shaped steel die (Larsson, 2008). The ring consists of many holes (channels) which the raw material are mechanically forced into with help of rollers (Larsson, 2008), see Fig 1.

Holm et al. (2006) has from material specific parameters such as sliding friction coefficient, elastic modules and Poisson ratio proposed a theoretical model that describes the pressure variation along the die channel. The model shows that the required pelletizing pressure increases exponentially with an increased die channel length. The model was verified experientially by Holm et al. (2007) with a single pelleter unit. The experiment showed that the back pressure (reverse pelletizing pressure) increased exponentially with increased die channel length. The theoretical model followed the same pattern as in the experiment. It should be noticed that the experiment was done on a single pelleter unit were the pelletizing pressure was controlled by the user, in contrast with a pellet mill where the pelletizing pressure is controlled by the material properties and specific die dimensions.

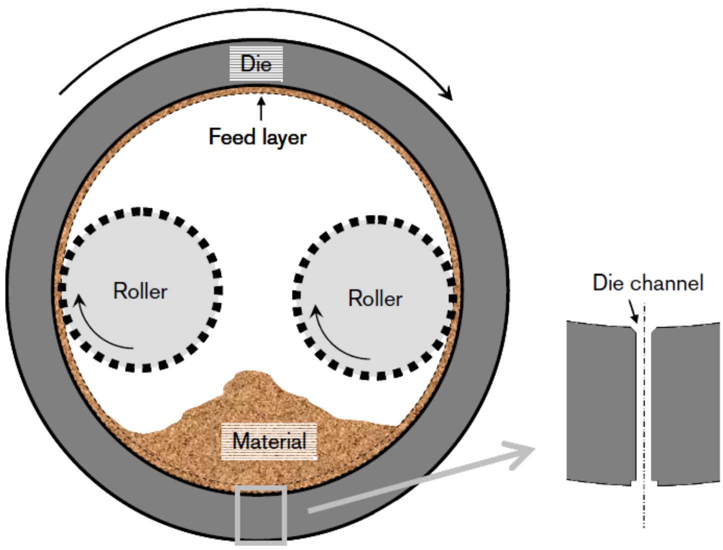

Fig 1. Schematic representation of a vertical pelletizer with a rotating die and two rollers, and enlargement of the die channel (Larsson, 2008).

4 Raw material properties

Pellet quality can be affected by storage time because a decrease of extractive content (Back, 1991; Larsson et al., unpublished; Bergström et al., 2010; Nielsen et al., 2010; Samuelsson et al., 2009; Filbakk et al., 2011a).

4.1 Storage time

Extractives are substances that can be extracted from the wood by organic solvents (Jirjis et al., 2006) and they consist of low molecular weight compounds such as fatty acids, waxes, sterols and terpenes, etc. (Back, 1991). A more precise appellation of extractives is oleophilic substances. When wood is freshly cut these substances are concentrated at the wood surface and creating a layer that prevents optimal binding between wood particles (Back, 1991). This phenomenon are by Back (1991) called deactivation but are by Stehr & Johansson (2000) called a Weak Boundary Layer (WBL). Active oxidation of this layer by solar radiation, ozone and fire is considered the most effective method to remove this layer (Back, 1991).

Assarsson (1969) showed that outdoor storage of Scots pine (*Pinus Sylvestris* L.) logs decreases the extractive content due to chemical auto-oxidation and microbiological activity. The process was dependent on temperature and available water and oxygen (Assarsson, 1969). The process is much faster in sawdust (Larsson et al., unpublished) and chipped wood then in stored logs on a landing site (Assarsson, 1969). In Scots pine logs the extractive content decreased from 3.6-2.6 % in 5 years of outdoor storage (Assarsson, 1969) compared to 3.7-1.8 % in 160 days stored Scots pine (*Pinus Sylvestris* L.) sawdust (Larsson et al., unpublished). Samuelsson et al. (2009) also showed a degradation of extractives during storage of Scots pine and Norway spruce (*Picea Abies* Karst (L)) sawdust for 140 days.

Back (1991) showed that the ability for oleophilic compounds to migrate to the wood surface is reduced if Scots pine logs are stored in three months. Back (1991) then assumed that the stored logs have a weaker layer and less amounts of oleophilic compounds compared to fresh cuts logs.

Bergström et al. (2010) and Nielsen et al. (2010) showed that removal of extractives result in pellets with higher compression strength. Nielsen et al. (2010) has a theory that the extractives prevent water to bind between the woody particles which resulting in low compression strength. Nielsen et al. (2010) suggest that the total amounts of extractive content are more important than the decrease of extractives that can be found after storage. Nielsen et al. (2010) found no large differences in compression, friction and pellet strength between a control and a storage group. The storage group was done in a heating cabinet at 60 °C for 21 days (Nielsen et al., 2010).

Bradfield & Levi (1984) has imitated whole tree assortments by adding different amounts of bark to the raw material. They concluded that when extractives and lignin contents exceeded 34 % the durability of pellets decreased.

A decrease (Samuelsson et al., 2009) or removal (Nielsen et al., 2010) of extractive content is increasing the energy consumption for pelletizing (Samuelsson et al., 2009; Nielsen et al., 2010). Both friction and compression increased when the extractives were removed (Nielsen et al., 2011). The authors suggest that extractives acts like lubricants in the pelletizing process and can decrease the energy consumption.

Stelte et al. (2011) studied inter-particle adhesion bounding in pellets of European beech (*Fagus sylvatica* L.), Norway spruce (*Picia abies* K.) and wheat straw (*Triticum aestivum* L.). In wood at a temperature of 20 °C Stelte et al. (2011) suggests that the bonding mechanisms were a combination of van der Waals forces and hydrogen boundings. In beech pellets and at a temperature of 100 °C there were also found solid bridges between adjacent particles, which results in pellets with higher durability. In wheat straw, Stelte et al. (2011) suggests that the bonding mechanisms was caused only by Van der Vaals forces due to high amounts of oleophilic compounds and low amounts of lignin which results in low durability pellets at both 20 and 100 °C.

Because storage time and extractive content are highly correlated many studies have been reported the effects of storage time for pellet quality. It have been shown that storage time is negatively correlated to bulk density (Filbakk et al., 2011a), fines (Samuelsson et al., 2009) and positively correlated to durability (Filbakk et al., 2011a; Samuelsson et al., 2009), and energy consumption (i.e. a degradation of extractives) and bulk density (Samuelsson et al., 2009).

5. Objectives

For further development of the Swedish pellet industry, the use of assortment of lodgepole pine (*Pinus contorta* Dougl. var. *latifolia*) is of particular interest mainly because of its high volume production. Whole tree and stemwood assortments can be milled and processed to fuel pellets but several properties have to be fulfilled to achieve high quality pellets. Pellet quality is a result of chosen raw material and its properties e.g. particle size distribution, ash content, storage time etc. and chosen process settings e.g. die channel length, die temperature, steam addition or drying, addition of binders. With respect to the European Standard EN: 14961-1: 2010 and EN 14961-2: 2011 this master thesis focus on the following pellet quality parameters; ash content (%), ash melting point (%), mechanical durability (%), amount of fines (%) and bulk density (kg/m^3). The energy consumption (A) was also monitored.

It is common known in the Swedish pellet industry that a storage period of sawdust increases the pellet durability. The reason for this are suggested being a decrease in extractive content and that the extractives prevent optimal particle-to-particle bounding (Bergström et al., 2010; Nielsen et al., 2010). Moisture content is a common known factor that increased bounding properties in wood particles (Grover & Mishra, 1996; Kaliyan & Morey, 2009; Mani et al., 2003). The research activities has not been active on different die channels lengths thus longer die channel lengths increase durability and energy consumption (Holm et al., 2006; Holm et al., 2007).

Therefore this thesis focus on how die channel length and raw material moisture content (process settings) and storage time (raw material property) effects pellet quality parameters stated above. The chosen intervals and experimental settings (Fig 2) were based on previous experiences on similar raw materials made at the Biofuel Technology Centre in Umeå, Sweden.

The objectives were to:

1) find suitable production window for process settings; i.e. to investigate which moisture content, die channel length and storage time that are suitable to obtain high quality pellets.

2) determine influence of process settings and storage time on pellet quality; i.e. to investigate how moisture content, die channel length and storage time affect pellet quality parameters bulk density, fines, mechanical durability and energy consumption.

3) determine influence of storage time on feedstock quality; i.e. to investigate how storage time of logs affects the ash content, ash melting temperature and extractive content.

using lodgepole pine stems (including bark) and whole trees (stems including bark, branches, tops and needles), respectively as feedstock.

6. Material and Methods

6.1 Biomaterial

Lodgepole pine (*Pinus contorta* Dougl. var. *latifolia*) trees were harvested at Långträsk, (latitude 65 °N, longitude 21 °E (x = 7247025, y = 1730287, RT90 coordinate system)), Norrbotten county, Sweden, in January 2011. Two different assortments were used in this study, stemwood (including bark) and whole trees (stems including bark, tops and branches with needles). Stand data are summarized in Table 2. Logs were delivered to the Biofuel Technology Centre, Swedish University of Agricultural Sciences in Umeå, Sweden for storage and pelletizing. The two assortments were put in two piles and stored on asphalt. The size of the piles after stacking was approximately sex meter wide and two meter high. The different storage times were counted from the day that the trees where harvested until the day of chipping. The first storage period was done under snow i.e. all 61 days for stemwood and the first 119 days for the whole tree assortment (SMHI, 2012), see Table 3.

Table 2. Stand data (before and after thinning) from the assortments stemwood and whole tree in Långträsk, Norrbotten County, Sweden.

	Before thinning		Thinned logs		
Assortments	Basal area (m^2/ha)	Stems / ha	Age (year)	Stem diameter (cm)[a]	Volume (m^3/ha)
Stemwood	25.3	1955	31	13.4	110
Whole tree	33.3	2085	31	13.9	139

a) Mean stem diameter at breast height.

Table 3. The three storage times for the assortments stemwood and whole tree.

Assortments	Days		
Stemwood	61[a]	242	325
Whole tree	132[a]	249	315

a) Winter storage under snow

6.2 Experimental design

The experimental design was developed using the software MODDE 9.0 (Umetrics AB, Sweden) and the same design was run both for whole tree and stemwood. The design consisted of three controllable factors at three levels; moisture content (9, 11 and 13 % of dry base weight), storage time (61, 242 and 325 days for stemwood and 132, 249 and 315 days for whole tree assortment see, Table 3) and die channel length (45, 50 and 55 mm). The original plan was to perform a Central Composite Face (CCF) centered design. However, because of misunderstandings in the performing procedure the design described in Fig 2 was used. Because of this, information was lost in the experimental settings that are marked with X in Fig 2.

Studied responses were fines (%), pellet bulk density (kg/m^3), mechanical durability (%) and pelletizer motor current (A). Achieved settings and measured responses for each pelletizing run are described in Tables 4 and 5, see section results.

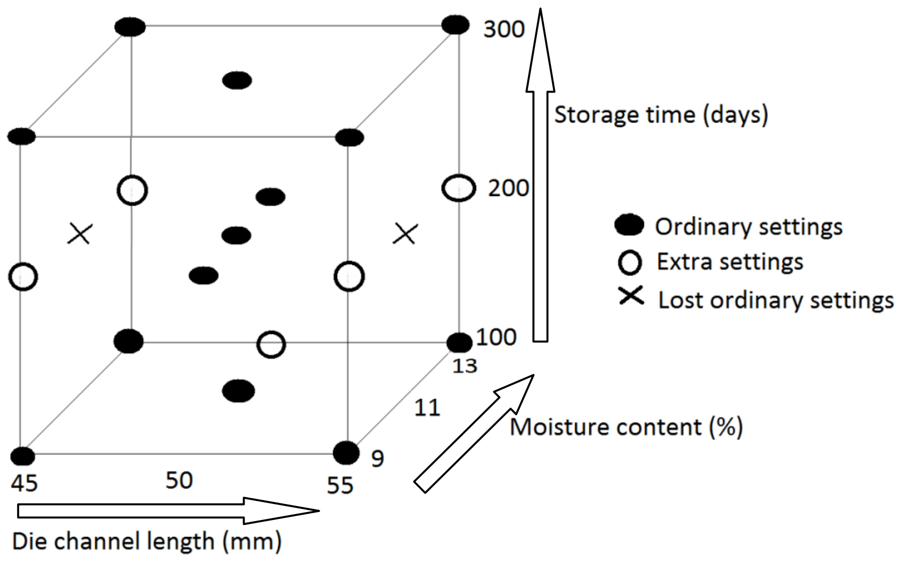

Fig 2. Both filled and empty circles are settings run in the experimental design. Settings marked with X should have been run (information losses). Empty circles were the “misunderstanding” trials.

6.3 Experimental setup

The materials were chipped with a tractor-driven wood chipper (Farmi CH 260, Farmi forest corporation, Idensalmi, Finland). After chipping, the materials were dried in a drying wagon using warm ventilation air (40-60 °C) until a desired moisture content of at least 7 % was reached. Dried chips were shredded (Lindner Micromat 2000, Linder-Recylingtech GmbH, Spittal, Austria) with 15 mm screen size, and hammer milled (Vertica Hammer DFZK-1, Bühler AG, Uzwil, Switzerland) with 4 mm screen size. Moisture content of 9, 11 and 13 % were obtained by adding water to the materials during mixing.

6.4 Pelletizing experiments

Pelletizing experiments were performed using a Bühler DPCB 420 pelletizer (Bühler AG, Uzwil, Switzerland) with a rotating die and a maximum softwood pellet production of approximately 500 kg/h. All experiments started with an initial warming up period (approximately 20 min) to achieve stable process and die temperatures before the sampling started. Pellet production rate was held constant at approximately 360 kg/h. For each experimental setting, sampling and measurements were done in triplicates. Each measurement period lasted for one minute, during which the amount of produced pellets were collected and weighed, milled raw material was sampled and corresponding process data were logged. Milled raw material was immediately sealed in plastic bags. Collected pellet samples were left overnight to cool down to temperatures of about 20 °C in open containers.

6.5 Experimental analyses

The amount of fines was measured manually by sieving approximately 6 kg of cold pellets (20 °C) through a 4 mm sieve. Fines were calculated as percentage loss of fine material in relation to the total sample weight. See Appendix A for formula.

Bulk density tests were determined according to the CEN standard (EN 15103: 2009). Bulk density was measured by filling a 5.4 liter bucket with pellets. The bucket was filled over the edge, dropped three times from a height of 15 cm and refilled after each fall. After the last fall, overflow pellets was removed with a rectangular piece of wood that were stroked over the bucket edges. See Appendix A for formula.

Mechanical durability tests were performed according to the CEN standard (EN 15210-1: 2009). Mechanical durability was tested by putting approximately 500 g of pellets in two boxes in a tumbling unit. The boxes were rotating total 500 laps in 10 minutes. After the treatment, the samples were sieved with a 3.15 mm sieve and the mechanical durability was calculated as the percentage of remained pellets in relation to the original weight. See Appendix A for formula.

Pelletizer motor current (A) was logged during the whole experiment period using the software Easy View Pro ver. 5.7.0.1 (Intab Interface-Teknik AB, Sweden). An average motor current value was calculated for each measurement period (one minute intervals).

Extractives were determined by using an extraction 4 system (Universal Extraction System B-811 from Büchi Labortechnik AG, Flawil, Switzerland). The extraction solvent consisted of a mixture of petroleum ether (boiling point: 40–60 °C) and acetone, with the volume-wise relation 90: 10. The extraction time was one hour and one pooled sample was taken from each triplicate. The method used is described by Arshadi & Gref (2005).

Ash content was analyzed according to the CEN standard (EN 14775: 2009) except that the raw material were milled with an 4 mm screen size (should have a particle size of 4 mm according to the standard. One pooled sample were taken from each triplicate. Seven samples were taken for each storage time except for the first storage period where only two samples were taken. In total 16 samples (Table 8) were taken. Approximately 2 g of milled raw material were put in crucibles and then placed in a drying cabinet that holds a temperature of 105 ± 10 °C for 16 hours. Thereafter, the samples were placed in an oven that holds a temperature of 550 ± 10 °C for 2.5 hours with a stirring of the samples after half-time. The ash content was thereafter calculated by the formula in Appendix A.

Ash melting behavior was analyzed by Bränslelabratoriet Umeå AB, Umeå, Sweden. The laboratory is accredited by SS-EN ISO/IEC 17025. The analyses were following the method ASTM D 1857-68 for ash melting points and done in oven (LECO AF 700). One pooled sample from each storage time was analyzed. A cone of ash were made and put in the oven. The temperature was successive increasing by 8 °C/min up to 1500 °C. When characteristic shape changes occur on the cone the temperature was registered. These changes were: Initial deforming temperature (IT) – the temperature when the first form changes occur on the top. Spherical temperature (ST) – the temperature when the cones height is equal to the base width. Half Spherical temperature (HT) – the temperature when the cones height is half of the base width. Melting point (MP) – the temperature when the cone has melted down to a height of 1.6 mm.

Moisture content was analyzed according to the CEN standard (EN 14774-2: 2009). A drying cabinet (1900x1000x820 mm) (Elvärmedetaljer AB, Sweden) calibrated to 105 ± 2 °C was used to dry the milled raw material in 750x600 mm trays for at least 12 hours. The samples were weighed with 0.1 g accuracy with balance (Mettler PM 4600 Delta Range, IT Instrument Teknik AB, Sweden) before and after drying. Moisture content was calculated by using the formula in Appendix A.

6.6 Modeling

Modeling of the responses was done in the software MODDE (Umetrics AB, Sweden) by using multiple linear regressions (MLR) of the factors. The factors were based on interactions and quadratic terms from storage time (days), raw material moisture content (%) and die channel length (mm). Response values were based on data from each measurement period. Each measurement period is based on averaged response values from the triplicates (duplicates for the excluded outliers).

Observations were considered as outliers if they deviated more than -4 and + 4 deleted studentized standard deviations from each response residual mean. The deleted studentized residual is the raw residual e_i divided by its standard deviation (s_i) where the standard deviation (s_i) is computed with observation (i) left out of the analysis, and corrected for leverage (MODDE, 2009). In both whole tree and stemwood assortment one measurement period was excluded because of unlikely raw material moisture content of 6 % (should have been 11 and 9 %, respectively). In stemwood one unlikely observation of bulk density of 813 kg/m^3 was excluded.

The Q^2 value is the percent of the variation of the response predicted by the model according to cross validation. Q^2 is a measure of how well the model predicts new data and can highest be 1 and lowest 0 (no predictive capability). The PRESS value are calculated by following formula from Myers (1986).

$$Q^2 = (SST\text{-}PRESS)/SST$$

with

$$PRESS = \sum_i \frac{(y_i - \hat{y}_i)^2}{(1-h_i)^2}$$

$$SST = \sum (y_i - \bar{y})^2$$

where for the ith object $\mathbf{y}_i$ is the observed response, $\hat{\mathbf{y}}_i$ the predicted response, $\mathbf{h}_i$ the ith diagonal element of the hat matrix $\mathbf{(X\,(X^TX)^{-1}\,X)}$ where ^{T} denotes the transposed matrix and **X** is each run in coded form from the calibration set. SST is the total sum of squared deviations in the response vector $\boldsymbol{y}$ from its mean $\bar{\boldsymbol{y}}$.

All factors were mean centered and range scaled from -1 to +1. This makes it possible to make comparisons between regression coefficients from different factors. The theory behind range scaling is more described by Myers & Montgomery (2002). The choice of ingoing factors in the models was chosen by maximizing the Q^2 value. This was done by excluding regression coefficients that were not significantly different from zero with a 95 % confidence interval or not increasing the Q^2 value. A regression coefficient can be non-significant but still enhance the Q^2 value.

As a validation measure the models were verified by calculating the root mean squared of cross-validation (RMSECV) by following formula:

$$RMSECV = \sqrt{\frac{\sum y_{obs-}y_{pred})^2}{n}}$$

Where y_{obs} were the observed responses, y_{pred} the predicted responses obtained from leave one-out cross-validation, and n was the number of observations in the test set. The RMSECV value is a measure of how good each response surface is i.e. the 4D contour plots (Fig 5-8).

6.7 Statistics

Statistical analyses of the regression coefficients were done in the software MODDE (Umetrics, Sweden). A significance level of 95 % was set in all statistic tests, both in MODDE and Minitab 16.1.

To examine whether the observations in the responses follow a normal distribution an Anderson-Darling goodness-of-fit statistic test was performed in Minitab (Minitab, Inc). If the p-value was >0.005 the data followed a normal distribution. To examine which type of transformation that was optimal a box-cox test was done in the MODDE. The box-cox test computes the best mathematical transformation.

Statistical analyses of ash and extractive contents for determining a difference between storage times were done in Minitab. A nonparametric Mann-Whitney test was done because the number of observations was low and equal variance was not present in the data.

6.8 Effect plots

For comparing the effect of the factors on each response, effect plots were conducted in MODDE. The effect plots have the advantage that is possible to estimate the effect of a factor along a gradient from its lowest value to its highest value. For example when the factor die channel length varies from 45-55 mm, the effect is representing the change in the response when the factor (component k) varies from -1 to 1 over its range, when all other factors are kept as in the reference mixture. See Appendix C for statistical notes.

7 Results

The results for each response variable are shown in Tables 4 and 5, together with experimental and achieved settings of the factors.

Table 4. Experimental settings and observed response values for stemwood.

Stemwood

Run	Exp. settings		Factors values			Responses values			
	Moisture (%)	Storage (days)	Moisture (%)	Storage (days)	Die channel length (mm)	Bulk density (%)	Durability (%)	Fines (%)	Motor current (A)
1	9	100	9.3	61	45	559.5	79.2	32.0	60.7
2	13	100	12.6	61	45	571.1	89.2	8.6	64.1
3	9	100	9.9	61	55	629.0	86.4	24.6	66.2
4	13	100	12.9	61	55	633.4	94.7	4.3	67.6
5	11	100	11.3	61	50	613.2	93.7	6.6	67.3
6[a]	13	100	12.9	61	50	632.6	95.1	3.0	67.5
7	11	200	11.5	242	50	666.9	95.3	3.9	72.2
8	9	200	9.4	242	50	642.3	93.9	5.8	70.1
9	13	200	13.5	242	50	652.1	96.6	2.0	70.4
10[a]	9	200	9.2	242	55	662.3	91.0	20.3	72.1
11[a]	13	200	12.6	242	55	662.9	95.6	5.6	73.4
12[a]	9	200	8.4	242	45	599.8	85.2	18.9	65.9
13[a]	13	200	11.9	242	45	622.4	93.7	8.5	67.3
14	11	300	11.4	325	50	640.1	92.4	5.2	68.4
15[b]	11	300	11.5	325	50	640.2	92.3	6.0	67.7
16[b]	11	300	11.5	325	50	640.7	92.5	6.9	67.6
17	9	300	10.0	325	55	640.9	86.6	18.7	73.8
18	13	300	14.3	325	55	653.3	96.7	2.7	74.6
19	9	300	9.8	325	45	595.4	79.2	40.4	63.7
20	13	300	13.7	325	45	651.1	95.0	3.8	68.2

a) Extra runs
b) Replicated runs

Table 5. Experimental settings and observed response values for whole tree.

Whole tree

Run	Exp. settings		Factors values			Responses values			
	Moisture (%)	Storage (days)	Moisture (%)	Storage (days)	Die channel length (mm)	Bulk density (%)	Durability (%)	Fines (%)	Motor current (A)
1	9	100	9.3	132	45	542.2	80.2	26.5	60.5
2	13	100	12.8	132	45	527.5	83.9	14.9	60.7
3	9	100	9.7	132	55	637.8	95.2	10.5	66.6
4	13	100	13.2	132	55	609.9	96.0	3.0	64.0
5	11	100	11.1	132	50	648.5	95.9	4.4	64.7
6[a]	13	100	12.8	132	50	620.6	95.9	2.6	66.0
7	9	200	9.5	249	50	644.6	92.7	14.4	70.4
8	13	200	13.3	249	50	652.0	96.6	4.1	70.4
9	11	200	10.9	249	50	658.6	95.2	8.8	70.3
10[a]	9	200	9.4	249	55	635.9	92.2	19.8	67.8
11[a]	13	200	13.4	249	55	656.3	95.9	7.0	69.8
12[a]	13	200	12.0	249	45	599.2	92.2	8.7	64.2
13[a]	9	200	9.7	249	45	578.3	83.3	26.9	63.8
14	11	300	11.4	315	50	586.2	84.6	27.2	64.0
15[b]	11	300	11.5	315	50	591.5	81.6	32.1	64.1
16[b]	11	300	11.3	315	50	596.3	88.1	21.2	65.6
17	9	300	10.4	315	55	634.0	89.6	19.7	71.1
18	13	300	14.4	315	55	659.5	88.4	12.3	71.1
19	9	300	9.5	315	45	568.3	81.5	35.1	64.0
20	13	300	13.7	315	45	625.3	92.2	13.4	66.8

a) Extra runs
b) Replicated runs

7.1 Model descriptions

Stemwood

All responses were modeled with scaled factors and the explained variation in the calibration set was equal or exceeding 90 % ($R^2 \geq 0.90$). The Q^2 values were from 0.78-0.88 which indicates good prediction accuracy of the response models. Most of the regression coefficients were significant ($p < 0.05$). Non-significant terms are marked in Table 6.

According to the Anderson-Darling test a transformation of data was required for durability and fines in the stemwood assortment. If the p value was <0.005 a transformation were conducted. The required transformation was detected from the box-cox test in MODDE. According to the box-cox test a negative log ($-{}^{10}\log(100-y)$) and log (${}^{10}\log(y)$) transformation was required for durability and fines, respectively (Table 6).

Whole tree

All responses were modeled with scaled factors and the explained variation in the calibration set was from 59-78 % (R^2). The Q^2 values were from 0.35-0.61 which indicate bad accuracy of the response models except for fines (0.61). Most of the regression coefficients were significant ($p < 0.05$). Non-significant terms are marked in Table 7.

Table 6. Summary of regression modeling coefficients for stemwood.

Stemwood				
Modell components	**Bulk density (%)**	**Durability[b] (%)**	**Fines[c] (%)**	**Motor current (A)**
Number of observations	20	20	20	20
F value	28.06	38.24	39.77	24.70
R^2	0.94	0.95	0.95	0.90
Q^2	0.78	0.87	0.88	0.78
RMSECV	6.88	0.95	3.37	1.11
Anderson-Darling: p value	0.032	<0.005	<0.005	0.346
Cofficents				
Constant	651.89	-0.66	0.61	70.28
Moisture content (MOI)	16.41	0.36	-0.56	1.89
Storage time (STO)	13.86	-0.01[a]	0.02[a]	1.67
Die channel length (DIE)	22.56	0.07	-0.04[a]	2.89
Interactions				
*STO*STO*	-24.48	-0.23	0.20	-3.20
*DIE*DIE*	-11.65	-0.16	0.37	
*MOI*MOI*		0.11[a]	-0.24	
*MOI*DIE*	-11.47			
*STO*DIE*	-9.09			0.87[a]

a) Non-significant term.

b) Transformed Neg Log: ($-{}^{10}\log(100-y)$).

c) Transformed Log: (${}^{10}\log(y)$).

Table 7. Summary of regression modeling coefficients for whole tree.

Whole tree				
Modell components	**Bulk density (%)**	**Durability (%)**	**Fines (%)**	**Motor current (A)**
Number of observations	20	20	20	20
F value	7.56	7.34	13.02	10.61
R^2	0.59	0.72	0.78	0.67
Q^2	0.35	0.42	0.61	0.49
RMSECV	24.43	2.91	4.61	1.88
Anderson-Darling: p value	0.210	0.022	0.330	0.075
Cofficents				
Constant	628.31	94.15	8.35	67.20
Moisture content (MOI)		3.98	-11.11	
Storage time (STO)	35.35	1.32[a]	2.72[a]	4.62
Die channel length (DIE)	32.73	4.51	-3.43	2.54
Interactions				
*STO*STO*	-64.56	-11.70	17.25	
*STO*DIE*		-4.39		
*DIE*DIE*				

a) Non-significant term.

7.2 MLR analysis and predication of the responses

Fig 3 and 4 shows the effect of the factors on the responses. The error bar is the 95 % confidence interval. Fig 5-8 show the predicted stemwood 4D contour plot values and show that optimum values was not obtained for all responses. Because of a bad accuracy (Q^2 value) for all responses in the whole tree model predicted 4D contour plots are not presented. Furthermore, RMSECV, Q^2 and R^2 values are shown in Tables 6 and 7.

7.2.1 Bulk density

Stemwood

The main factors for bulk density were the squared term of storage time (STO*STO) and die channel length (DIE), see Fig 3 A. A Q^2 value of 0.78 indicates an approved prediction accuracy of the model which is further supported by a RMSECV value of 6.88 and an explained variation R^2 value of 0.94 (Table 6). The 4D contour plot of predicted bulk density (Fig 5) shows that bulk density increases with moisture content and die channel length. Furthermore, bulk density is highest in die channel 55 mm, 130-280 storage days and moisture content >10 %.

Whole tree

The two main factors that affect bulk density where the squared term of storage time (STO*STO) and the storage time, see Fig 4 A. A Q^2 value of 0.35 indicates a bad prediction accuracy of the model and is further supported by an explained variation R^2 value of 0.59 (Table 7).

7.2.2 Durability

Stemwood

The main factors for durability were the moisture content (MOI) and the squared term of storage time (STO*STO), see Fig 3 B. A Q^2 value of 0.87 indicates a good prediction accuracy of the model which is further supported by a RMSECV value of 0.95 and an explained variation R^2 value of 0.95 (Table 6).The 4D contour plot of predicted durability (Fig 6) shows that durability increases with moisture content and are highest in die channel length 50 mm, storage time 200 days and moisture content >11 %.

Whole tree

The main influential factor for durability was the squared term of storage time (STO*STO), see Fig 4 A. A Q^2 value of 0.42 indicates a bad prediction accuracy of the model. The explained variation R^2 value was 0.72 (Table 7).

7.2.3 Fines

Stemwood

The main factor that affected fines was the moisture content (MOI), see Fig 3 C. A Q^2 value of 0.87 indicates good prediction accuracy of the model which is further supported by a RMSECV value of 3.37 and an explained variation R^2 value of 0.95 (Table 6). The MLR-models for durability and fines are shown to be highly correlated. The regressions coefficients are almost the same but opposite to each other. High moisture content results in high durability and low amounts of fines, respectively (Fig 3). The 4D contour plot of predicted fines (Fig 7) shows that fines decreases with increased moisture content and are lowest in die channel 50 mm, storage time of 200 days and moisture content >11 %.

Whole tree

The main influential factors for fines were the squared term of storage time (STO*STO) and moisture content (MOI), see Fig 4 C. A Q^2 value of 0.61 indicates an approved prediction accuracy of the model which is further supported by a RMSECV value of 4.61 and an explained variation R^2 value of 0.78 (Table 7). The MLR-models for durability and fines are shown to be correlated. The regressions coefficients are almost the same but opposite to each other. High moisture content results in high durability respective low amounts of fines.

7.2.4 Energy consumption

Stemwood

The two main factors that affects energy consumption were the squared term of storage time (STO*STO) and die channel length (DIE), see Fig 3 D. A Q^2 value of 0.78 indicates approved prediction accuracy of the model which is further supported by a RMSECV value of 1.11 and explained variation R^2 value of 0.90 (Table 6). The 4D contour plot of predicted stemwood energy consumption (Fig 8) shows that energy consumption increases with increased moisture content. Furthermore is energy consumption highest at 150-300 storage days, die channel length of 55 mm and moisture content ≥12 %.

Whole tree

The main influential factors for energy consumption was the squared term of storage time (STO*STO) and storage time (STO), see Fig 3 D. A Q^2 value of 0.49 indicates bad prediction accuracy of the model which is further supported by an explained variation R^2 value of 0.67 (Table 7).

7.3 Comparing regression coefficients

A comparative study between regression coefficients were made for the two assortments. A summary of the three largest factors that affect the responses are shown in Table 8.

The squared term of storage time (STO*STO) was the most important factor that influenced energy consumption and bulk density but it also affected the durability and fines to some degree. The exception for the whole tree assortment was that the squared term of storage time was the most important factor that influencing durability. The large influence of the squared term of storage time explained the similar pattern in the stemwood 4D contour plots (Fig 5-8) i.e. that around 200 days gave the highest values for bulk density, durability and fines.

Die channel length was one of the most important factor for stemwood that influenced bulk density and second factor that explained energy consumption.

Moisture content was the most important factor for stemwood that influence durability and fines, but also affected energy consumption and bulk density to some degree. The exception for the whole tree assortment was that moisture content did not affect energy consumption and bulk density significant.

Table 8. Summary of regressions coefficients between stemwood and whole tree. Numbers are the effect of each regression coefficients in a decreasing scale, from 1 to 3. Bold numbers indicates similarities between stemwood and whole tree assortments.

	Factors						
Responses	Moisture	Storage	Die	STO*STO	DIE*DIE	MOI*MOI	STO*DIE
Stemwood							
Bulk density	3		2	**1**			
Durability	1			2	3		
Fines	1				2	3	
Energy consumption	3		2	**1**			
Whole tree							
Bulk density		2	3	**1**			
Durability			2	1			3
Fines	2		3	1			
Energy consumption		2	3	**1**			

7.4 Stemwood effect plots

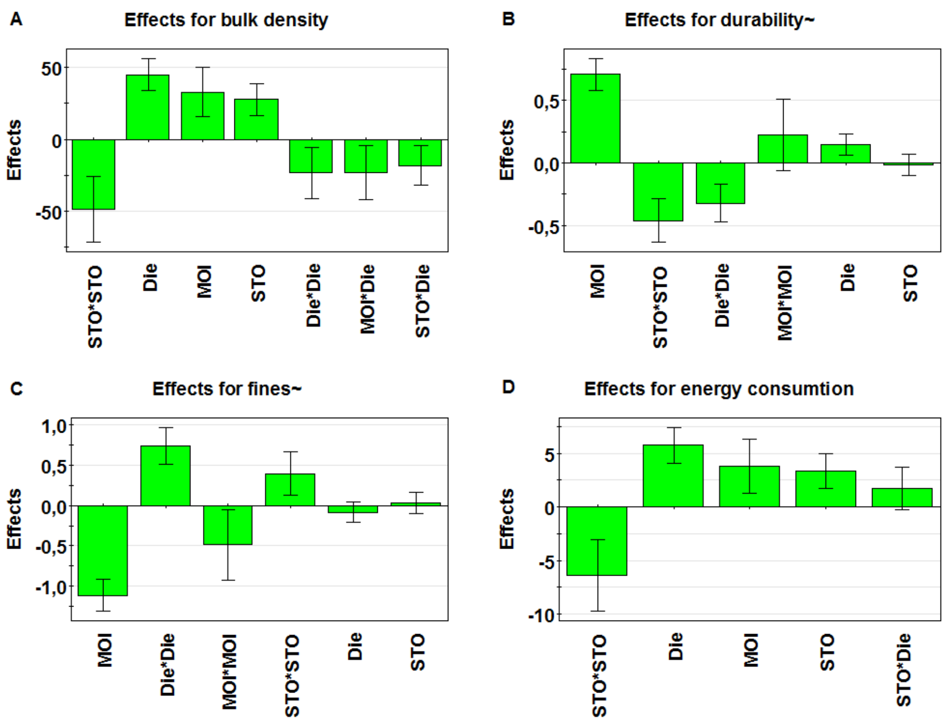

Fig 3. Main effects that describe the responses for stemwood. Error bar is the 95 % confidence interval. Transformed responses (durability and fines) are marked with a ”~” symbol in each figure heading.

7.5 Whole tree effect plots

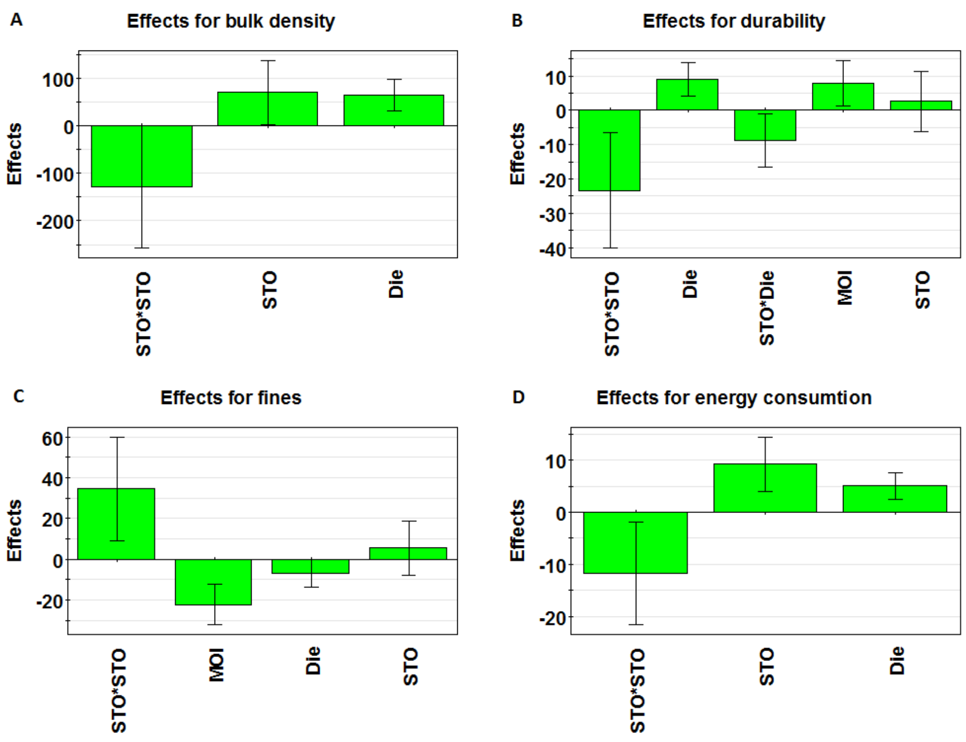

Fig 4. Main effects that describes the responses for whole tree. Error bar is the 95 % confidence interval.

7.6 Stemwood 4D contour plots

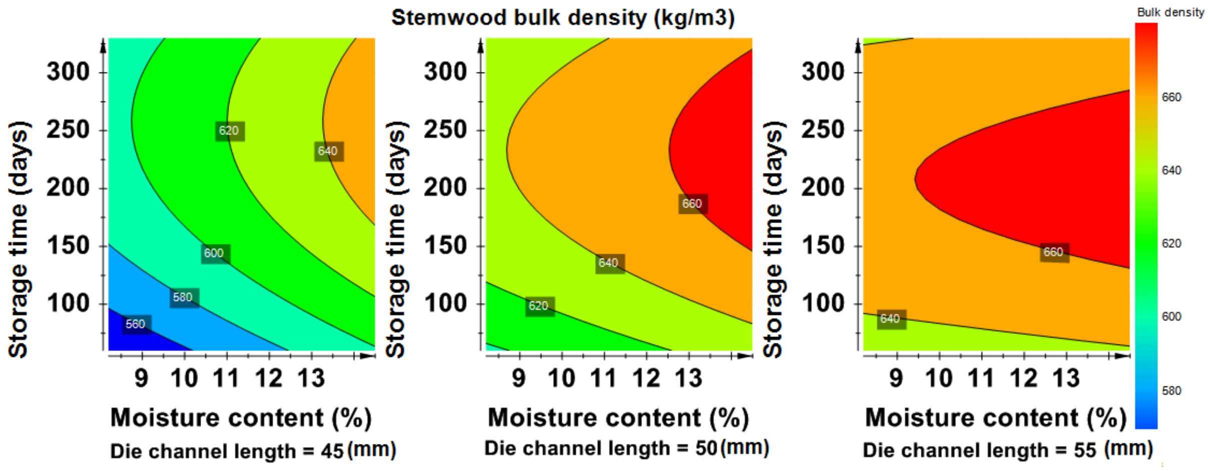

Fig 5. Bulk density for different storage times (days), raw material moisture contents (%) and die channel lengths (mm).

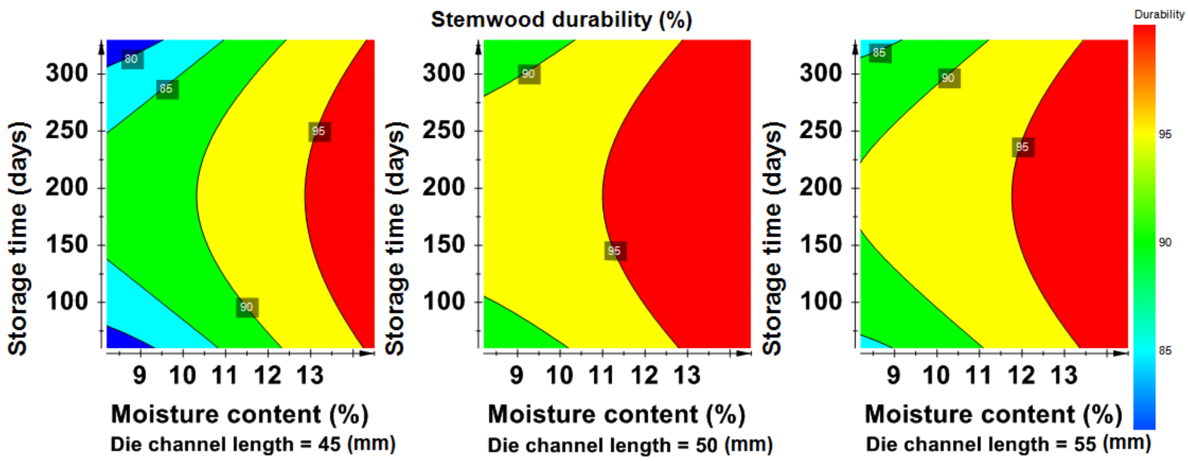

Fig 6. Durability for different storage times (days), raw material moisture contents (%) and die channel lengths (mm).

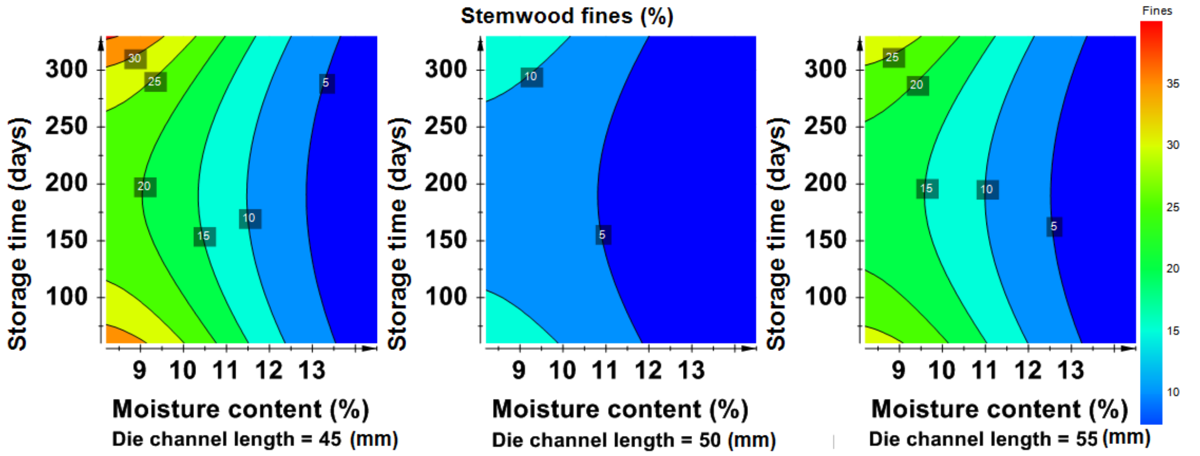

Fig 7. Amount of fines for different storage times (days), raw material moisture contents (%) and die channel lengths (mm).

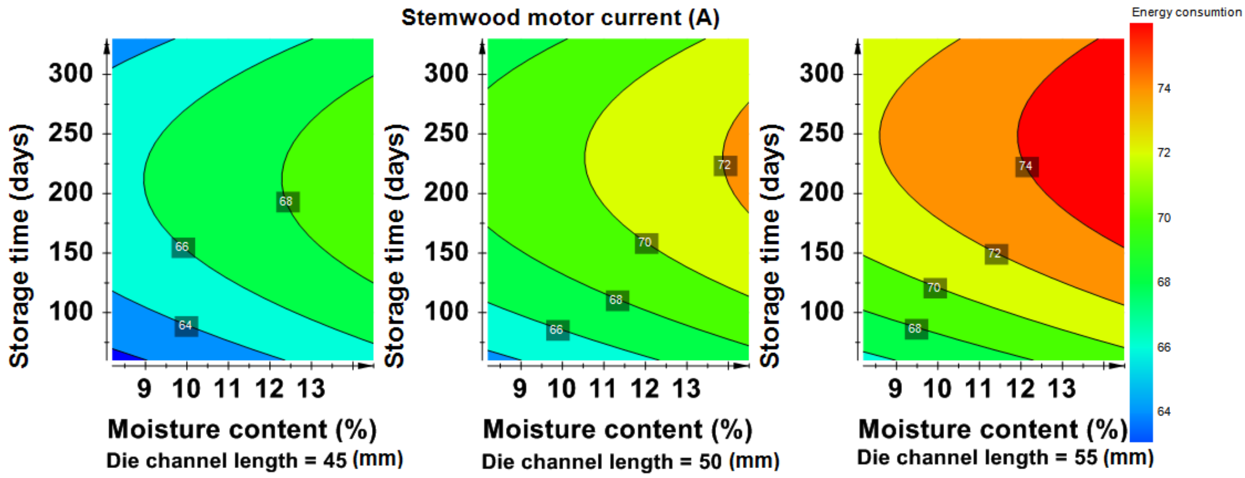

Fig 8. Energy consumption for different storage times (days), raw material moisture contents (%) and die channel lengths (mm).

7.7 Stemwood production window

To find the production window for high quality pellets a multiple response optimization was performed on the factors storage time (days), moisture content (%) and die channel length (mm). According to the highest pellet quality class in the CEN Standard (EN 14961-2: 2011) the bulk density should be ≥600 kg/m^3, durability ≥97.5 % and fines ≤1 %. The sweet spot plot in Fig 9 shows the intervals when these criterions are met according to conducted MLR-models. Low energy consumption (under the mean of 68 A) and the criteria of ≤1 % amount of fines did not occur together with the other criteria and therefore not placed in the sweet spot plot shown in Fig 9.

The sweet spot plot (Fig 9) shows that high quality pellets were achieved with die channel length 50 mm, whole storage period and moisture content >13.8 %. Furthermore, high quality pellets was achieved in die channel 55 mm, storage time from 100-290 days and moisture content >13.8 %.

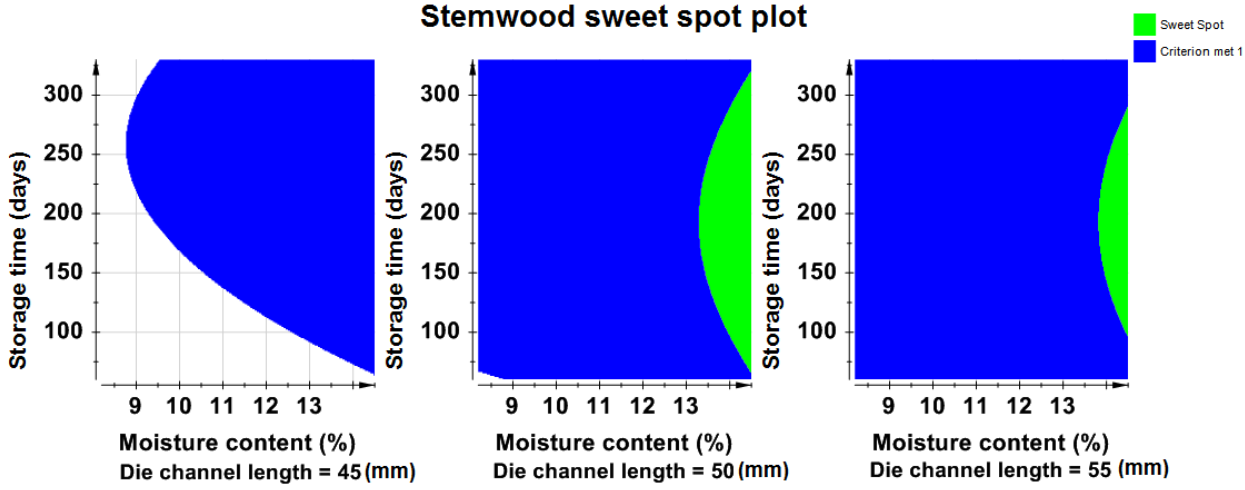

Fig 9. Sweet spot plot that shows in which intervals the assortment stemwood have bulk density ≥600 kg/m^3 and durability ≥97.5 % for different storage times (days), raw material moisture contents (%) and die channel lengths (mm). Criterion met 1 (blue) = bulk density >600 kg/m^3. Sweet Spot (green) = bulk density >600 kg/m^3 and durability >97.5 %).

7.8 Ash content

Ash content varied from 0.41-0.44 % (mean 0.42 %) for stemwood and from 0.58-0.64 % (mean 0.61 %) for whole tree assortment. The number of observations was too low for storage time 61 days in stemwood and for 132 days in whole tree assortment to make a significance difference test between storage times. The other storage times were not significant diffrent from each other (p-value 0.0736), see Table 9.

7.9 Extractives

Extractive content varied from 2.0-2.9 % for stemwood (mean 2.6 %) and from 3.9-4.3 % (mean 3.9 %) for the whole tree assortment. The number of observations was too low for storage time 61 days in stemwood and for 132 days in whole tree assortment to make a significance difference test between storage times. A significant decrease (p-value 0.0033) was found between storage time 242 and 325 days for stemwood. The extractive content decreased from 3.0-2.0 %. No significant difference (p-value 0.0967) was found in the whole tree assortment, see Table 9.

Table 9. Average ash and extractive contents for three different storage times and the two assortments. Numbers of observations (n) are presented before the average values.

Assortsments and storage time (days)	n	Ash (%)	p-value[a]	n	Extractives (%)	p-value[a]
Stemwood						
61	2	0.41 (0.03)		2	2.9 (0.1)	
242	7	0.42 (0.02)	0.0736	7	3.0 (0.5)	0.0033
325	7	0.44 (0.03)		7	2.0 (0.4)	
Mean		0.42			2.6	
Whole tree						
132	2	0.61 (0.02)		3	4.3 (0.1)	
249	7	0.58 (0.06)	0.0736	7	3.4 (0.5)	0.0967
315	7	0.64 (0.03)		7	3.9 (0.4)	
Mean		0.61			3.9	

Standard devation in brackets

a) p-value between the storage times 242 and 325 & 249 and 315 days.

7.10 Ash melting temperatures

The ash melting temperatures for stemwood assortment was >1500 °C for all characteristic shape changes of the ash cone. For whole tree assortment with storage time 132 days the temperatures were 1490 °C and for the other storage times 1500 °C, see Table 10.

Table 10. Charterteristic shape changes (IT, ST, HT, MT) was registered and temperatures in °C were logged for different storage times and assortments.

Assortments and storage time (days)	IT	ST	HT	MT
Stemwood				
61	>1500	>1500	>1500	>1500
242	>1500	>1500	>1500	>1500
325	>1500	>1500	>1500	>1500
Whole tree				
132	1490	1490	1490	1490
249	1500	1500	1500	1500
315	1500	1500	1500	1500

IT) Intial deforming temperature

ST) Spherical temperature

HT) Half spherical temperature

MT) Melting point

8. Discussion

Durability

One of the most important pellet quality parameter is the durability of pellets (Kaliyan et al., 2009) and moisture content is greatly affects pellet durability (Grover & Mishra, 1996; Obernberger & Thek, 2010; Kaliyan & Morey, 2009; Filbakk et al., 2011a; Mani et al., 2003; Nielsen et al., 2009; Samuelsson et al., 2009; Rhen et al., 2005; Lehtikangas, 2001).

In this study the durability increased with increased moisture content and was highest when moisture content was over 13 % (Fig 6). An increasing pattern between durability and moisture content is in accordance to other studies (Filbakk et al., 2011a; Larsson et al., 2008; Lehtikangas, 2001). However the opposite pattern have also been reported for pellet strength (Nielsen et al., 2009), compression strength (Rhen et al., 2005) and durability within studied intervals (Samuelsson et al., 2009).

An explanation for this can be that water acts as a binding agent that increases the area of connection for Van der Waal forces (Kaliyan & Morey, 2009; Grover & Mishra, 1996; Mani et al., 2003) and at the same time water can sorb to hydrogen bounding sites and therefore occupying the site for particle-to-particle bounding (Nielsen et al., 2009). The result indicates that the optimum value has not been found because durability still increases with increased moisture content. In future research the moisture content should be increased to find were the maximum value appears. It should be noticed that the maximum value between durability and moisture content differs between chosen raw material and process settings (Filbakk et al., 2011a).

Another important factor that have been reported to affect pellet durability is the extractive content (Larsson et al., unpublished; Bergström et al., 2010; Nielsen et al., 2010; Samuelsson et al., 2009; Filbakk et al., 2011a). A significant decrease in extractive content was found when storage time was increased from 242 to 325 days for the stemwood assortment (Table 9), but the result is unsure because only seven samples were investigated. The average extractive content varied from 2.0-3.0 % for stemwood (Table 5). Koch (1996) has reported an average extractive content of 2.87 % in lodgepole pine (*Pinus contorta* Dougl. var. *latifolia*) entire stemwood. A decrease of extractives has been shown after storage of Scots pine (*Pinus sylvestris* L.) logs (Assarsson, 1969; Back, 1991), Scots pine and Norway spruce (*Picia abies* K.) sawdust (Samuelsson et al., 2009) and pellet durability has increased after removal (Bergström et al., 2010; Nielsen et al., 2010) or by decreasing the extractive content (Samuelsson et al., 2009).

The result showed that durability increased when high extractive contents appear at storage times of 150-250 days (Fig 6 and Table 9). Due to low number of samples this result is unsure. However, this is opposite to results reported by Bergström et al. (2010) and Nielsen et al. (2010) were high amounts of extractives resulted in low durability. The authors suggest that extractives substances form a “Weak Boundary Layer” that prevents water to bind between the wood particles and therefore affect durability negatively. Nielsen et al., (2010) also mentioned that the total amounts of extractive content are more important than the decrease in extractives that can be found after storage. Nielsen et al. (2010) found small differences in compression strength, friction in the die channel and pellet strength between a control and a storage group that was stored in a heating cabinet at 60 °C for 21 days (Nielsen et al., 2010).

Therefore, based on the Nielsen et al. (2010) results the effect of storage and thereby extractive content may not have a crucial role in pellet strength and durability. But if all extractives are removed, pellet strength (Nielsen et al., 2010) and compression strength (Bergström et al., 2010) are greatly increased. Jirjis et al. (2006) further suggest that the bounding abilities can be explained by the chemical composition of lignin, hemicellulose and extractives. Bradfield & Levi (1984) states that if the extractives and lignin contents exceeded 34 % the durability of pellets decrease.

Bulk density

The bulk density should be as high as possible because it increases the energy content. In this study bulk density increased with longer die channel lengths and was highest in die channel of 55 mm (Fig 5). This is possibly caused by the fact the raw material have a longer distance to travel in the die channel and therefore increases the friction and compression, resulting in higher back pressure and thus, denser pellets.

Bulk density increased with increasing moisture content (Fig 5). Many studies have shown the opposite, namely that bulk density should decrease with moisture content (Larsson et al., 2008; Samuelsson et al., 2009; Filbakk et al., 2011a). The explanation for this can be that lodgepole pine has a higher optimum value for moisture content for maximum bulk density. In future research the moisture content should be increased to find were the maximum value appears.

The 4D contour plot for bulk density (Fig 5) gives a clear indication that the die channel lengths should be longer than used in this study since the optimal values have not been found for bulk density. No reference could be found were bulk density or energy consumption was increasing with moisture content.

Energy consumption

Energy consumption affects the overall costs of pellet production and should be minimized. Energy consumption was increased with increasing moisture content (Fig 8). These findings are in opposite to Samuelsson et al. (2009), Filbakk et al. (2011a), Nielsen et al. (2009) and Arshadi et al. (2008) where Samuelsson et al. (2009) suggests that moisture lowers the friction in the die channel and therefore decreases the energy consumption.

Energy consumption was highest when storage time was 242 days and when extractive content was highest (3.0 %) (Fig 8 and Table 9). Due to small number of samples, these results are unsure. However these findings are in opposite to Samuelsson et al. (2009) and Nielsen et al. (2010) where it is suggested that extractives acts like plasticizers and lubricants that lower the friction in the die channel and therefore decrease the energy consumption.

A longer die channel length increased the energy consumption (Table 9). Previous studies have shown that required pelletizing pressure increases exponentially with increased die channel length (Holm et al., 2006; Holm et al., 2007). With higher needed pelleting pressure it is natural that the energy consumption is increased and therefore a longer die channel results in higher energy consumption.

Ash content

The ash content in pellets is an important quality factor because high ash contents can create problems in burners due to ash formation elements that lead to severe slagging (Lindström et al., 2010), lower combustion efficiency and higher emissions of particles (Werkelin et al., 2005; Ericsson, 2006).

The average ash contents for stemwood varied from 0.41-0.44 %. Koch (1996) reported an average ash content of 0.26 % in lodgepole pine (*Pinus contorta* Dougl. var. *latifolia*) entire stemwood. The ash contents in whole tree varied from 0.58-0.64 % so the ash content was aproximatly 1.4 times higher in the whole tree assortment (Table 9). No reference were found on average ash contents in lodgepole pine whole tree assortments, but generelly bark and needles have higher ash contents compared to its stemwood (Petersson & Nordfjell, 2007). Lindström et al. (2010) showed that a whole tree assortment of Scots pine (*Pinus sylvestris* L.) had 2.2 times higher ash contents compared to its stemwood.

However, the results in this thesis indicate that both stemwood and whole tree assortments pass the ash content requirements (<0.7 %) for high quality pellets stated in the European standard EN 14961-2: 2011. This fact means especially that stemwood of this diameter class does not have to be barked which reduces costs in processing and labor.

Limitations in study and model

The predicted production window in Fig 9 indicates that an area for production of high quality pellets are found, but it should be noticed that the area is based on only one observation in the green area in die channel 50 and 55 mm, respectively. The replication run 15 and 16 (Table 4 and 5) were run after each other which reduce the ability to compute a standard error measure of each experimental setting. A RMSECV value was instead calculated for the response surface (Table 6 and 7).

The analyses in extractive and ash content has an error range of 10 % and 5 %, respectively (Samuelsson pers. comm. 2012).

Effects for the industry

The results of this thesis can be applied on three main markets; forestry, pellet and energy industry. Since optimal values of all examined responses could not be found within the range settings chosen for this study, due to low moisture content and short die channel lengths, it is difficult to make any clear guidelines for the industry, but based on the results the following recommendations can be made: die channel length ≥50 mm, logs stored for 200 days (61 days under snow) and raw material moisture content >13.8 % before pelletizing (Fig 9). But it should be noticed that storage of logs did not increase pellet quality substantially.

Pellet industry

The results indicate that high moisture content is crucial in the production of high quality pellets (Fig 5-8). If high quality is desired, the moisture content should be over 13 % before pelletizing (Fig 9). These results mean that huge energy savings can be made because the raw material needs not to be dried below 13 % in moisture content.

Results also showed that die channel length of 50 mm or longer should be used. On the other hand durability is highest for die channel 50 mm (Fig 9). A longer die channel increases energy consumption (Fig 8) but will result in higher quality of pellets (Fig 9). Especially the bulk density will increase if die channel of 55 mm is used (Fig 5).

A storage time of logs in approximately 200 days (61 days under snow) results in higher quality pellets (Fig 5-8). This means that storage of logs must be done at some place in the chain from forest to industry. But the results showed that storage did not have a crucial role for pellet quality. The gain in higher bulk density, durability, fines and lower energy consumption probably does not pay the higher cost related to storage of logs.

Forest industry

If stemwood will be used as a feedstock in pellet production the price of stemwood must be higher than the pulpwood assortment. The trend is that the price of biomass will increase as the demand for renewable resources is increasing. Intensive cultivation of lodgepole pine can be one opportunity for producing raw material for the pellet industry as well as for the heat and power plants because of the trees high volume production. In Sweden lodgepole pine has on average 36 % higher volume production than the domestic Scots pine (*Pinus sylvestris* L.) independent of site index (Elfving et al., 2001).

Energy industry

Thermal power plants strive to find a raw material with as high energy value as possible with good combustion properties. The result shows that both stemwood and whole tree assortments of lodgepole pine can be combusted with low risk for ash melting (Table 10) or high amounts of ash contents (Table 9). This result can be applied not only for the non-industrial pellet market but also for combustion of chipped whole tree assortments of lodge pole pine in thermal power plants. It should be noticed that the ash content will be higher if only forest residues are combusted.

9. Conclusions

1) According to the highest pellet quality class A1 in the CEN Standard (EN 14961-2: 2011) a production window with high quality pellets were found with following settings: die channel length 50 mm, whole storage period (61-325 days) and moisture content >13.8 % (Fig 9). Furthermore was high quality pellets achieved with die channel 55 mm, storage time 100-290 days and moisture content >13.8 % (Fig 9). The results showed that high energy consumption occur with high bulk density and durability. It seems that the study have investigated a too narrow interval in moisture content and die channel length. Fig 9 clearly indicates that optimum settings are not found within the studied intervals for each factor. Thus, additional experiments must be conducted where especially higher moisture content and longer die channels should be included. This conclusion is also supported by the results in the 4D contour plots (Fig 5-8).

2) Moisture content was the most significant factor explaining the durability and the proportion of fines and the relationships were positive and negative, respectively within the studied interval of 9-13 % (Fig 3). The relationship between storage time and all responses were positive (up to 200 days) with the exception of energy consumption (Fig 3). Die channel length was one of the most significant factors that affected the bulk density and energy consumption and the correlation was positive (Fig 3). Energy consumption and durability were highest when storage time was 242 days and when extractive content was highest (3.0 %) (Fig 8 and Table 9).

3) Mean ash content in stemwood were 0.42 % and 0.61 % in whole tree assortment (Table 9) which results that the ash content passes the requirments (<0.7 %) for high quality pellets stated in the European standard EN 14961-2: 2011. Mean extractive content were 2.6 % in stemwood and 3.9 % in whole tree assortment (Table 9). Ash melting temperatures was approxmentely 1500 °C for both stemwood and whole tree assortments (Table 10) which indicate good combustion properties. Storage time did not affect the ash content sigificantly (Table 9).

The overall conclusion is that stemwood (including bark) of lodge pole pine within the found production window can be used as feedstock to produce fuel pellets of high quality and that whole tree assortment could not be predicted with high predication accuracy.

Appendix A

<u>Fines</u> (%)

$$\text{Proportion of fines} = \frac{(m_f - m_e)}{m_f}$$

m_f = Wight before sieving (kg)
m_e = Weight after sieving (kg)

<u>Bulk density</u> (kg/m^3)

$$\text{Bulk density} = \frac{m_p}{V_P}$$

m_p = Weight of pellets (kg)
V_P = Volume of pellets (m^3)

<u>Mechanical durability</u> (%)

$$\text{Mechanical durability} = \frac{m_e}{m_f}$$

m_f = Weight before sieving (kg)
m_e = Weight after sieving (kg)

<u>Moisture content</u> (%)

$$\text{Moisture content} = \frac{m_2 - m_3}{m_2 - m_1} * 100$$

m_1 = Weight of the empty tray (g)
m_2 = Weight of the tray with moist sample (g)
m_3 = Weight of the tray with dry sample (g)

<u>Ash content</u> (%)

$$\text{Ash content} = \frac{(m_3 - m_1)}{(m_2 - m_1)} * 100 * \frac{100}{(100 - m_{ad})}$$

m_1 = empty tray (g)
m_2 = tray and raw material (g)
m_3 = tray and ash (g)
m_{ad} = Moisture content of sample (%)

Appendix B

Stemwood

Variable	Mean	StDev	Minimum	Maximum	Range
SBulk density	630,45	29,43	559,51	666,87	107,36
SDurability	91,23	5,29	79,21	96,74	17,54
SFines	11,39	10,81	2,03	40,41	38,38
SEnergy consumtion	68,440	3,579	60,680	74,553	13,873

Whole tree

Variable	Mean	StDev	Minimum	Maximum	Range
WBulk density	613,62	38,97	527,53	659,45	131,92
WDurability	90,06	5,69	80,18	96,59	16,41
WFines	15,62	9,99	2,63	35,12	32,49
WEnergy consumtion	66,301	3,326	60,537	71,143	10,607

Appendix C

From MODDE 9.0 help section.

Screening plots

When the objective is to find the component effects on the response, the coefficients of the Cox reference linear model are directly proportional to the Cox effects. The Cox effect is the change in the response when component k varies from 0 to 1 along the Cox axis. That is the axis joining the reference point to the $\mathbf{k^{th}}$ vertex.

Effect plot

The effect plot displays the adjusted Cox effects. The adjusted effect of component **k** is:

$k = r_k * t_k$
$r_k = U_k - L_k$

$t_k = b_k/(T-s_k)$

where:

r_k is the range of factor k

t_k is the total Cox effect

T is the mixture total. In most cases T=1.

b_k is the unscaled uncentered coefficient

s_k is the value of the factor at the reference mixture

The **Effect Plot** is only available for screening designs using the Cox model.

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