Effects of Extractives on the Physical Characteristics of Scots Pine Sawdust Fuel Pellets

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Abstract

In order to evaluate the effect of extractives on particle bonding, pellets were produced from extracted and nonextracted Scots pine sawdust in a laboratory piston press pelletizer. In the experiment, the raw material and process parameters were fresh (nonextracted) and acetone-extracted sawdust, sawdust moisture content (6% and 12%), piston pressure (70 and 150 MPa), and press temperature (100°C and 180°C). The resulting pellets were evaluated and compared for density, compression strength, and moisture sorption. The relationship between factors and responses was evaluated by partial least squares regression. In the present study, it was found that pressure and temperature had a positive effect on both pellet density and compression strength. Extracted sawdust gave pellets with a higher density and compression strength than pellets made from nonextracted sawdust. Moisture sorption between the produced pellets showed no significant differences. Results of this study provide a plausible explanation for why pellets produced from stored sawdust with low amounts of extractives have better strength properties than pellets produced from fresh sawdust.

Wood pellets are produced mainly from milled wood sawdust with a relatively wide particle size distribution (Bergström et al. 2008). In the pelletizing process, sawdust particles are forced together and densified under heat and pressure in the die holes to form cylindrical compact (dense) pellets. Moisture is necessary in the process because it facilitates the heat transfer and favors particle bonding (Rhén et al. 2005). In addition to raw material moisture content, particle size, die pressure, and die temperature are important parameters in pelletizing (Krug et al. 1978, Rhén et al. 2005). No adhesives are used in the process, and pellets should therefore be considered as binderless particle composites (Rhén et al. 2005). Wood pellets are easy to transport, possess uniform size, and have high energy density and low moisture content, and therefore are suitable for small heating plants and furnaces (Bhattacharya et al. 1989).

There are several explanations proposed for the mechanisms by which particles in binderless wood composites, including pellets (Nielsen et al. 2009), are bonded, such as by thermal softening of lignin (Goring 1963), pectin bonding (Pickard et al. 1961), bonding through hemicellulose degradation products (Mobarak et al. 1982), or by mechanical particle interlocking (Li and Liu 2000) in which size, shape, and surface roughness of the wood particle influence the bonding strength (Gardner 2002).

The influence of raw material extractives on pellet characteristics such as density, compression strength, and moisture sorption is not well understood. However, during the manufacture of particleboards (hardboard), it has been observed that active sites on the particles may be blocked by lipophilic substances that have migrated to the particle surface during drying and hot pressing, thus obstructing wettability and reducing bond strength (Back 1987, Gardner 2002). It may be assumed that similar hydrophobic extractive migration to the surface occurs as well during sawdust drying and pelletizing, thus affecting particle bonding in the pellet (Nielsen et al. 2010). This view is supported by the fact that pellets pressed from fresh sawdust compared with those pressed from stored sawdust show differences in durability. Pellets pressed from stored sawdust or from a mixture of fresh and stored sawdust generally have higher durability and bulk density than

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pellets pressed from entirely fresh sawdust (Lehtikangas 2001, Samuelsson et al. 2009). During the storage of sawdust (maturation), lipophilic extractives are broken down through microbiological and auto-oxidative processes (Hemingway et al. 1971, Arshadi and Gref 2005, Samuelsson et al. 2009), resulting in increased particle surface wettability and improved particle bonding (Back 1987, Gardner 2002). Stronger bonding between the particles will most likely minimize the springback after compression, thus resulting in higher density (Nielsen et al. 2010). Knowledge about the influence of extractives on pellet physical characteristics could make it possible to fine-tune the raw material handling process in pelletizing industries, resulting in less variation in pellet quality.

Based on the above given assumption, the objective of this work was to compare pellet density (PD), compression strength (CS), and moisture sorption (MS) of pellets pressed from extracted and nonextracted (fresh) Scots pine (*Pinus sylvestris* L.) sawdust. The pellets were individually produced in a laboratory-scale piston press under strictly controlled conditions.

Experimental

Raw material

Fresh, newly ground and dried Scots pine stem wood sawdust was collected at a pellet plant in Luleå, northern Sweden. At delivery, the sawdust had a moisture content (MC; % of fresh weight) of 9 percent. The particle size distribution of the sawdust was measured by sieving, and the dry weight (DW) of the different fractions was calculated (Table 1).

Raw material preparation

About 200 g of fresh sawdust was sampled and soaked twice with 1 liter of acetone in a capped beaker for 72 hours at room temperature as described by Yasuda et al. (1998). The mixtures were filtered, and the filtrate was evaporated to dryness on a rotary evaporator and weighed. The yield of the acetone-soluble compounds was 4.0 percent of sawdust DW. The dried acetone-soluble compounds were further extracted with petroleum ether. The solvent was evaporated, and the petroleum ether–soluble compounds were weighed. The yield of petroleum ether–soluble lipophilic compounds was 3.7 percent of sawdust DW. The extracted sawdust residue was air dried for 24 hours at room temperature in a fume hood and then for 48 hours at 50°C to a constant weight and kept in a sealed container until pelletizing. About 500 g of fresh sawdust was also sampled from the same sawdust batch, dried, and kept as above. Subsequently, the materials (fresh and extracted sawdust) were conditioned to an MC of 6.0 and 12.0 percent by adding water in aerosol form. Samples were then equilibrated for 48 hours in a cold-storage room.

Table 1.—Raw material particle size distribution of fresh Scots pine sawdust.

Particle size (mm)	Share of dry weight (%)
<0.25	13.8
≥0.25 to <0.5	20.1
≥0.5 to <1.0	50.2
≥1.0 to <1.4	12.6
≥1.4 to <2.0	3.2
≥2.0 to <2.8	0.1
Total	100

Pelletizing

Pellets from fresh and extracted sawdust were individually produced in a laboratory-scale piston press with a die diameter of 12.0 mm and under strictly controlled pressure, temperature, and pressing time (Rhén et al. 2005). Pellets were produced at 70 and 150 MPa, 100°C and 180°C, and for a pressing time of 10 seconds. For each pellet, 2.0 g of sawdust was used. The newly produced pellets were stored in airtight containers at room temperature until testing. Each parameter combination of the pelletizing process was replicated eight times, and a total of 128 pellets were pressed (Table 2).

PD and CS

Each pellet was measured for weight and volume, and the single PD was calculated. The CS was measured by compressing the pellet radially between two parallel plates moving together at a speed of 0.4 mm/min (Rhén et al. 2005) until the pellet collapsed. During compression, the required force (N) was registered. CS was defined as the quota between the maximum forces registered during compression and the length of the pellet (N per millimeter). Three pellets from each trial of pellets were tested for CS.

MS

The MS test was performed at SCA Packaging Research Laboratory in Sundsvall, Sweden, according to a method used for moisture uptake measurement in paper. The MS test was performed by placing pellets in a moisture chamber for 24 hours to equilibrate at a relative humidity (RH) of 30 percent and a temperature of 23°C. Subsequently, the RH was increased to 85 percent for 48 hours. The weights of the pellets were measured at the initial state (equilibrium) and then after 1, 2, 3, 4, 5, 6, 7.5, 24, 28, 31.5, and 48 hours. The MS was defined as the relative increase in weight compared

Table 2.—Raw material and process parameters of pellet trials, each replicated eight times.

	Raw material parameters		Process parameters	
Trial	Type of sawdust	Moisture content (%)	Pressure (MPa)	Temperature (°C)
P1	Fresh	6	70	100
P2	Fresh	6	150	100
P3	Fresh	6	70	180
P4	Fresh	6	150	180
P5	Fresh	12	70	100
P6	Fresh	12	150	100
P7	Fresh	12	70	180
P8	Fresh	12	150	180
P9	Extracted	6	70	100
P10	Extracted	6	150	100
P11	Extracted	6	70	180
P12	Extracted	6	150	180
P13	Extracted	12	70	100
P14	Extracted	12	150	100
P15	Extracted	12	70	180
P16	Extracted	12	150	180

with the initial weight directly after pelletizing. Two pellets per trial were sampled for measuring of MS.

Statistics and regression analysis

The relationship between the factors (raw material, MC, pressure, and temperature) and the responses (PD, CS, and MS at 30% and 85% RH) was evaluated by partial least squares (PLS) regression. The software used for the regression and statistical calculations was MODDE 8.0.2.0 (Umetrics Inc., Kinnelon, New Jersey). All significance tests were done at the 95 percent level. R^2 is the fraction of the variation of the response explained by the model, and Q^2 is the fraction of the variation of the response predicted by the model according to cross-validation. Values close to 1 for both R^2 and Q^2 indicate a very good model with excellent predictive power. The design gives the regression coefficients (b_0, b_1, . . . , b_5) in a model relating the factors (x_1, x_2, . . . , x_5) to a response y:

$$y = b_0 + b_1x_1 + b_2x_2 + \cdots b_5x_5 + e$$

Results and Discussion

Statistics and regression analysis

A summary of the PLS model statistics and the regression coefficients is shown in Table 3. Three principal components were found significant by cross-validation. No interaction or quadratic factors were included in the model. At high raw material MC (12%) and low press temperature (100°C), durable pellets could not be produced. This led to some missing values in the experimental design (experiment trials P5, P6, P13, and P14). Despite the missing values, it was possible to make a good PLS model from the remaining experiments.

PD and CS

The PD ranged from 1.04 to 1.18 g/cm^3 (Table 4). Of the process parameters, both pressure and temperature had a positive effect on the PD. This is in accordance with previous published works (Rhén et al. 2005, Kaliyan and Vance Morey 2009). In the present study, pressure had a somewhat higher effect. At high raw material MC, the PD decreased. This observation is also supported by published works (Rhén et al. 2005, Samuelsson et al. 2009). Extracted sawdust gave a higher PD than fresh sawdust, although the effect of extractives was smaller than for the other factors but still significant (Table 3; Fig. 1, left).

Table 3.—The three-component PLS model overview[a]

	PD (g/cm^3)	CS (N/mm)	MS	
			30% RH	85% RH
Prediction				
R^2	0.841	0.880	0.900	0.891
Q^2	0.813	0.864	0.782	0.828
Factors			Coefficients	
Constant	1.105	37.204	46.317	8.417
Fresh	−0.011	−1.448	−0.051 (NS)	−0.016 (NS)
Extracted	0.011	1.448	0.051 (NS)	0.016 (NS)
Moisture content	−0.028	−8.787	0.474	−0.221
Temperature	0.024	5.748	2.172	0.402
Pressure	0.030	2.672	0.086 (NS)	−0.021 (NS)

[a] Values are for the explained variation (R^2) and the predicted variation (Q^2) for the individual responses (PD, CS, and MS at 30% and 85% RH) and for scaled and centered coefficients of the model. PD = pellet density; CS = compression strength; MS = moisture sorption; RH = relative humidity; NS = not significant.

Table 4.—Properties of tested pellets.[a]

Trial	PD (g/cm^3), $n = 8$	CS (N/mm), $n = 3$	MS (%), $n = 2$[b]	
			30% RH	85% RH
P1	1.06 (0.03)	31.86 (3.21)	41.1 (1.2)	52.0 (1.1)
P2	1.11 (0.01)	33.75 (1.07)	41.6 (0.9)	52.6 (0.9)
P3	1.11 (0.01)	42.51 (2.91)	48.4 (1.3)	62.0 (1.5)
P4	1.17 (0.01)	53.08 (1.83)	47.8 (0.2)	61.0 (0.3)
P7	1.04 (0.02)	26.51 (1.24)	48.9 (0.7)	61.4 (0.6)
P8	1.08 (0.02)	27.72 (1.08)	49.1 (0.7)	61.5 (0.6)
P9	1.06 (0.01)	34.75 (2.93)	42.0 (0.5)	53.3 (0.6)
P10	1.14 (0.01)	40.81 (1.64)	42.5 (0.7)	53.9 (0.8)
P11	1.11 (0.01)	40.99 (0.97)	47.8 (0.3)	61.0 (0.4)
P12	1.18 (0.01)	53.20 (1.67)	47.7 (0.1)	60.8 (0.1)
P15	1.07 (0.01)	30.39 (1.22)	49.3 (0.5)	61.9 (0.8)
P16	1.13 (0.02)	30.88 (1.64)	49.8 (1.1)	62.3 (1.0)

[a] Values are averages with standard deviations in parentheses. PD = pellet density; CS = compression strength; MS = moisture sorption; RH = relative humidity; n = number of replicates.
[b] Values were taken at equilibrium (24 h) for 30 percent RH and at 48 hours from equilibrium for 85 percent RH.

The CS ranged from 26.51 to 53.20 N/mm (Table 4). Of the process parameters, both pressure and temperature had a positive effect on the pellet CS. In this study, temperature had a significantly greater effect. A high MC of the sawdust lowered the CS of the produced pellets and was the most influencing factor for this property. Extracted raw material gave higher CS than fresh raw material, although the effect was smaller than for the other factors but still significant (Table 3; Fig. 1, right).

These observations in PD and CS support the hypothesis that lipophilic substances block active sites on the raw material particles (Back 1987, Nielsen et al. 2009), resulting in minimized springback after compression, which is in accordance with findings in the Nielsen et al. (2010) study. Results from industrial pelletizing have also shown that prolonged sawdust storage time influences pellet bulk density positively (Samuelsson et al. 2009).

In Figure 2, CS as a function of PD for all trials (average values) is shown. When density increased from 1.04 to 1.18 g/cm^3 (13.5%), the CS increased about 92 percent (Fig. 2). The process factors had equal effects on the CS as they had on the PD of the pellets.

MS

Most of the MS occurred in the first part of the test at 30 percent RH. In this stage, the relative MC increased by about 46 percent-units for all pellets tested and ranged from 41.1 to 49.8 percent. In the second stage at 85 percent RH (48 h), an additional increase in relative MC of about 8 percent-units was detected and had increased to between 52.0 and 62.3 percent with an average value of 58.6 percent (Table 4). No significant difference in MS between pellets pressed from fresh sawdust and those from extracted sawdust could be detected in the present study. High pressing temperature resulted in drier pellets with increased MS (Fig. 3).

However, MC of the sawdust influenced the MS in different ways at 30 and 85 percent RH. At 30 percent RH, a high initial MC of the raw material gave a higher MS, but at

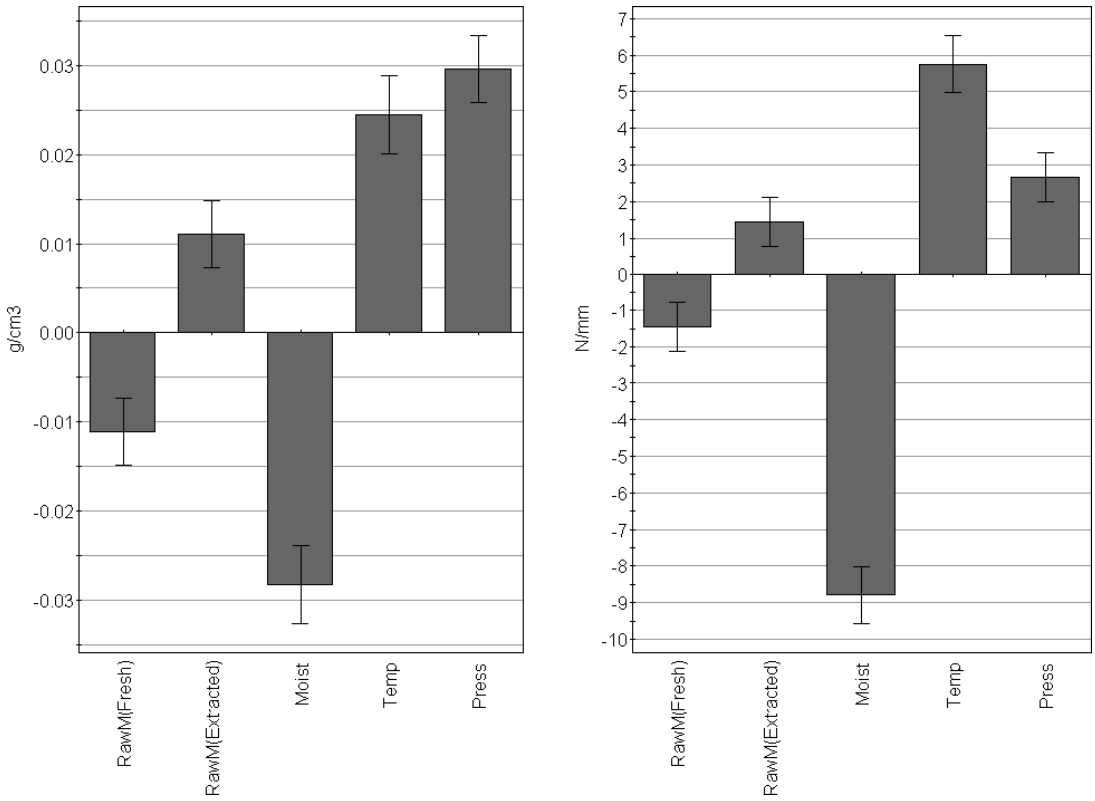

Figure 1.—Scaled and centered regression coefficients for the responses single pellet density (left) and compression strength (right).

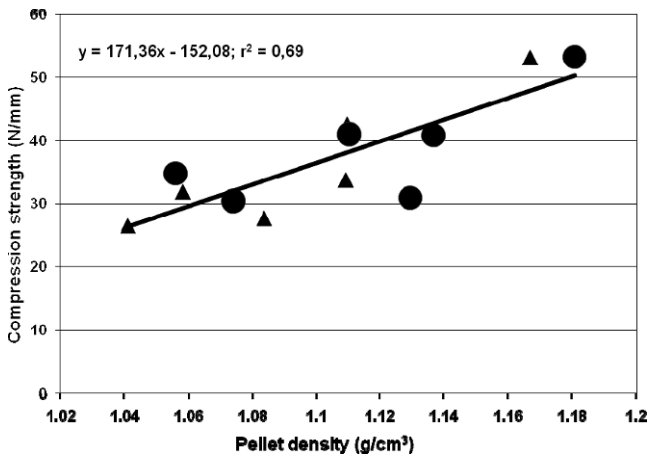

Figure 2.—Compression strength as a function of pellet density. Circles and triangles indicate pellets made from extracted and fresh (not extracted) sawdust, respectively.

85 percent RH, a high initial MC of the raw material resulted in a lower MS (Fig. 3). This can be explained by the very low initial MC of the pellets produced at high temperatures. The effect of the initial sawdust MC on MS in pellets could not be explained in this study.

Conclusions

The results obtained from this study demonstrate that a high temperature and high pressure in the pelletizing process give stronger and denser pellets. A high initial MC of the sawdust give pellets with lower density and CS compared with a low MC. This is in accordance to previous studies. Pellets pressed from extracted sawdust results in higher CS than pellets pressed from fresh sawdust. The reason for this might be that removal of extractives results in better interparticle contact during the compaction and therefore stronger bonding. An increase in density was also observed for pellets pressed from extracted sawdust. Results of this study explain why pellets produced from stored sawdust with low amounts of extractives have better strength properties than pellets produced from fresh sawdust. The MS properties of the pellets are not influenced by the extractives content of the sawdust. These properties are most likely closer associated to the temperature in the pressing stage and on the initial MC of the material entering the press.

Acknowledgments

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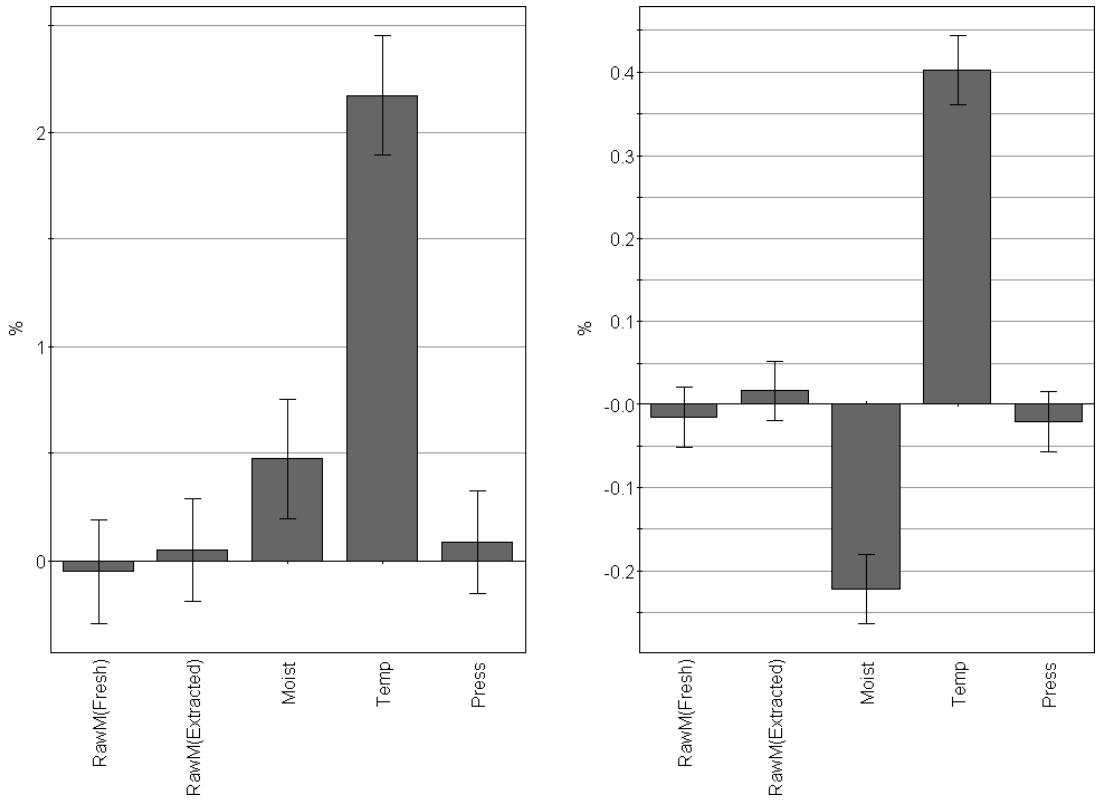

Figure 3.—Scaled and centered coefficients for the responses moisture sorption at 30 percent RH (left) and 85 percent RH (right).

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