

PELLETISING INVASIVE SHRUBS-DENSIFYING WITH DIFFERENT TIGHTENING STRENGTHS AND ITS INFLUENCE ON PELLETS QUALITY

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Abstract. Samples of *Acacia dealbata*, an invasive species, and of *Salix atrocinerea*, a draft summers resilient species, were collected and processed in order to be pelletized in a conventional low duty machine, static axis rollers/rotating die. The object of pelletizing is to increase the bulk density, ensuring a dense and cohesive product that meets international standards, namely concerning durability and water resistance. The mechanical properties involved depend on processing parameters, which induce chemical and mechanical phenomena among wood particles; of those undergone during pellets production only a few were completely studied. The mechanical strength of pellets is an important parameter since it is the base for good durability; friability, an indirect measure of the level resistance to withstand breakage, is a sensitive factor during handling and transportation.

The process of producing pellets from *Acacia dealbata* and *Salix atrocinerea* was initiated with predefined sizes harvesting, solar drying samples to 12-15% humidity, wet basis, milling the material and sorting it into distribution sizes that resulted in adequate average size.

In order to evaluate the specific energy used in the pelletizing process, temperatures and relative humidity of the atmosphere inside the pelletiser machine were collected, as well as the temperatures attained by the cast iron base and cover and by the axis where the rollers were assembled; the shaft was instrumented using two MTS extensometers; also, the level of the material inside the tub, above the die, was controlled. The contact forces between the rollers, the wood and the die, taking place during the extruding process, were calculated, and an average of 17 kN was obtained. All the above data were acquired with National Instrument LabView 8.6 and NI USB-6008 DAQ, Picolog Recorder using a TC-08 USB datalogger and Catman, Spider8-30.

Phenomena that are inducing an increase on the compressive strength of pellets were approached by microscopy showing strong bonding on the particles interface. Durability resistance tests were performed to assess the compliance to the international standard of 97.5%.

Keywords: bioenergy; invasive shrubs; pellets; extrusion; friability

1. INTRODUCTION

Vegetable biomass is the most important renewable energy source. It is also a major contribution to reducing greenhouse gas emissions and replacing fossil fuels (Faaij, 2006). Given the existence of raw material in Portugal and the negative externalities (forest fires) of letting this raw material untreated in forests, the National Forest Strategy (DGRF, 2006) proposes that priority should be given to further utilisation of that resource. Australian species of the genus *Acacia* are an environmental problem in several areas of Europe, especially in France, Spain and Portugal, due to the invasive character of some of them (Richardson and Rejmánek, 2011; Marchante *et al.*, 2008; Campos *et al.*, 2002). In Portugal, these species are not allowed in plantations and the restrictive law recommends their control or eradication through large scale plans (MADRP, 2005; Santos *et al.*, 2004). Silver wattle (*Acacia dealbata* Link.), which has been reported to have an „extremely high invasive potential” (Wilson *et al.*, 2011) is one of the most difficult species of the genus to eradicate, due to its resprouting ability from stump and roots, and persistence of seed bank that allow easy reestablishment after fire or clear cut (Sanz *et al.*, 2004); in addition, this species allows occasional small profits as provider of leafy twigs and flowers for ornamental purposes (Fernandes, 2008). On the other way, the versatile nature of biomass as a fuel enables it to be utilized everywhere, but at the same time makes biomass a complex and difficult fuel (Khan *et al.*, 2009).

The quality of the pellets is dependent on the pressure values used to compact the biomass, on the temperature, on the water content and on the particle size distribution (Lestander *et al.*, 2012). Kaliyan and Morey (2010) concluded that the softening of the biomass natural binders using moisture and temperature in the range of glass transition is important to make durable particle-particle bonding. Many experiments have been made and papers published with the experiments

based on simple compression with or without a heating element (Dhamodaran and Afzal, 2012; Kaliyan and Morey, 2009).

Biomass comprises natural macromolecular compounds. Some primary components are cellulose, hemicellulose, lignin, starch, proteins, lipids, simple sugars, water, hydrocarbons, ash, and other compounds. The ratio of the main structural components of biomass, cellulose, hemicellulose and lignin, is approximately 55:20:25% (Kitani, 1999). Lignin, a natural biopolymer which has historically been considered an unwanted product during paper and pulp manufacturing (Hatakeyama and Hatakeyama 2010), is a natural binder, whose presence is important in the final pellets quality as it would result in improved mechanical properties (Stelte *et al.*, 2011a; Sanadi and Caulfield, 2008).

Based on SEM microscopy, Stelte *et al.* (2011b) have observed the fracture surface of pellets prepared from different biomass types in order to study the bonding mechanisms that hold the pellet particles together. They produced pellets from beech (hardwood type), spruce (softwood type) and straw (grasses) at two different temperatures, 20 and 100 °C. At 20°C all pellets presented poor mechanical strength and was justified by a weak bonding between pellet particles. For wood pellets, the bonding is a combination of van der Waals forces and H-bonding, while the presence of wax on the straw surface resulted in poor adhesive strength (dominated by van der Waals forces) ascribed to a weak waxy boundary layer. Pellets produced at 100°C presented a greater mechanical strength. The higher values were obtained for beech pellets in which were observed solid bridges between adjacent particles that were ascribed to the lignin softening and inter-diffusion. However, spruce did not show solid bridge formation, which was attributed to the fact that the higher glass transition temperature of softwoods lignin did not allow sufficient lignin to flow. In this case, H-bonding and van der Waals forces (and some mechanical interlocking) are the primary mechanism of bonding.

This work assesses the durability and quality (mechanical resistance) from pellets made in an actual industrial machine. Two different type of pellets were used, *Acacia dealbata* (Silver wattle) and *Salix atrocinerea* (Grey willow), manufactured specifically for this work. Silver wattle is a hardwood species that has fast growing characteristics and, in some European countries, is legally defined as invasive; Grey Willow is a low commercial value (in what concerns the traditional industrial sectors- paper wood pulp and furniture) fast growing hardwood species. Therefore, both could be envisaged as alternative to other conventional wooden resources for pelletizing.

2. MATERIALS AND EXPERIMENTS

Samples of *Acacia dealbata* and *Salix atrocinerea* species were collected in a local forest, dried in a solar kiln, monitoring the moisture content. The drying process was carried out in the summer and each batch took 5 days on average. A hammer mill, model GKLC-19PK2010, grounded the raw material and the product output passed through a 6 mm screen. The mean diameter after screening with a AS200 Retsch shaker was 793 µm for *Acacia dealbata* and 601 µm for *Salix atrocinerea* samples. The sieving, for each sample, was of 10 minutes with oscillation amplitude of 1.50 mm.

3. EXPERIMENTAL SETUP

3.1 Pelletization

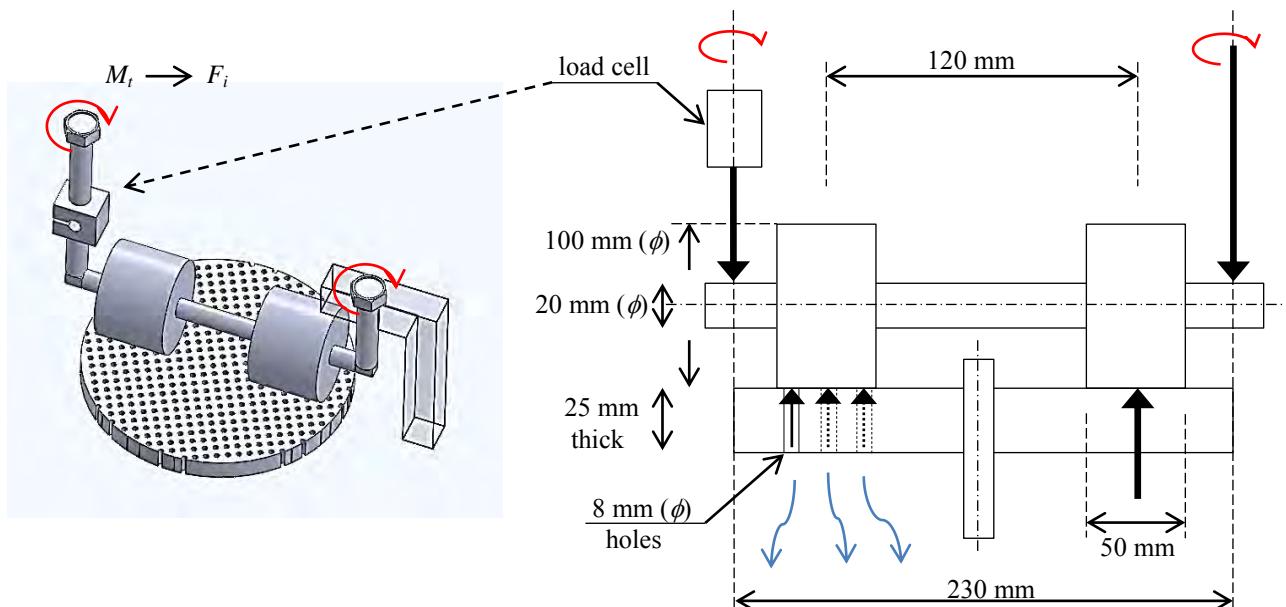


Figure 1. Experimental set-up schemes, pelletizer machine.

The pelletizer machine is a low cost fixed rolls (rolling on a static axis) flat die rotating base, through which the pellets are extruded. The machine, a 15 kW, 200 kg/h AGP GK5500 Pellet Mill, was modified to suit research needs, namely changing the dimensions and the shape of the hopper and replacing the primitive cylindrical roller bearings by more resistant elements, both to strength and temperature, as well as using different tightening screws and new heavy duty self-locking nuts. Additionally, an experimental setup was put in place specifically to serve the purpose of measuring the forces that were being used in the pelletizing process (Fig.s 1 and 2).

A complete set of variable acquisition data, online, was added. The different temperatures (roller axis, upper and lower tub), ambient and the inner hopper air relative humidity values were acquired, as well as the force values taking place during the pellets manufacturing process. All the samples used were made in the machine of Fig.s 1 and 2. In Tab. 3 a general description of their physical properties can be examined.



Figure 2. Experimental set-up, pelletizer machine: load cells and axis adjustments.

Newton's Second Law,

$$\sum F = m \frac{d^2x}{dt^2} \quad (1)$$

combined with Hooke's Law, that gives the force exerted by an ideal spring, F_s , as

$$F_s = -kx \quad (2)$$

where k is the spring constant, gives the following equation (Eq. 3) for a mass-spring problem:

$$m \frac{d^2x}{dt^2} + kx + F_i = F_c \quad (3)$$

with m the mass subjected to the force F_c , F_i the initial force and x the displacement from the natural length, measuring the change from the equilibrium position at time t .

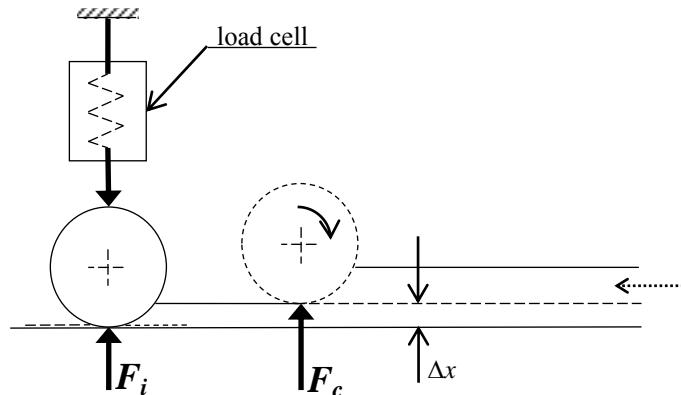


Figure 3. Initial (static) and final deformation of the plate/load cell set.

Considering that the inertial term in Eq. (3), $m \frac{d^2x}{dt^2}$, is small compared with the compression force value, the force balance becomes,

$$kx + F_i = F_c \quad (4)$$

Therefore, the compression force, given by the sum of the initial clamping force with the force exerted by the material introduced into the space between the roller and the plate, is equal to the force measured by the load cell.

The initial force applied is the result of the initial, static, tightening torque imposed to both the threaded shafts that are acting on the rollers axis. That initial static force, applied when the machine is turned off, causes a deformation of the plate/load cell set, designated by Δx_i .

The initial force F_i , a minimum for the operating conditions, is therefore

$$F_i = k_s \times \Delta x_i \quad (5)$$

where k_s is the spring constant of the plate/load cell set. Once the pelletizing machine is turned on, the material will begin being fed and it will be pressed by the rollers against the die; before an orifice arises, there will be an increase in the acting force, a function of Δx ,

$$F_c = k_s \times \Delta x + F_i \quad (6)$$

where k_s is the (same) set spring constant. The total force exerted reaches a maximum for a maximum displacement Δx_{max} , corresponding to the final thickness of the material being pressed between the roller and the die (Mediavilla, Fernández and Esteban, 2009):

$$F_{max} = k_s \times \Delta x_{max} \quad (7)$$

This maximum force value is variable as it depends, among others, on the average particle size, biomass density, bulk value presented and mass flow rate at which the material it is being fed into the pelletizing machine. Moreover, variations of shape and orientation of fibers and flaws result in a large scattering range of strength data (Li *et al.*, 2000). When the manufacturing machine was operating, average constant temperature periods were identified and, when temperature stabilized, the output pellets were collected for a minimum of thirty seconds.

3.2 Pellets mechanical testing

Mechanical strength is an important parameter for a mechanized use of a solid fuel. Single pellet strength measurements have been accepted as a good method for assessing rupture strength values. Also, the goal was comparing values between pellets made from different initial load conditions, more than determining an exact value.

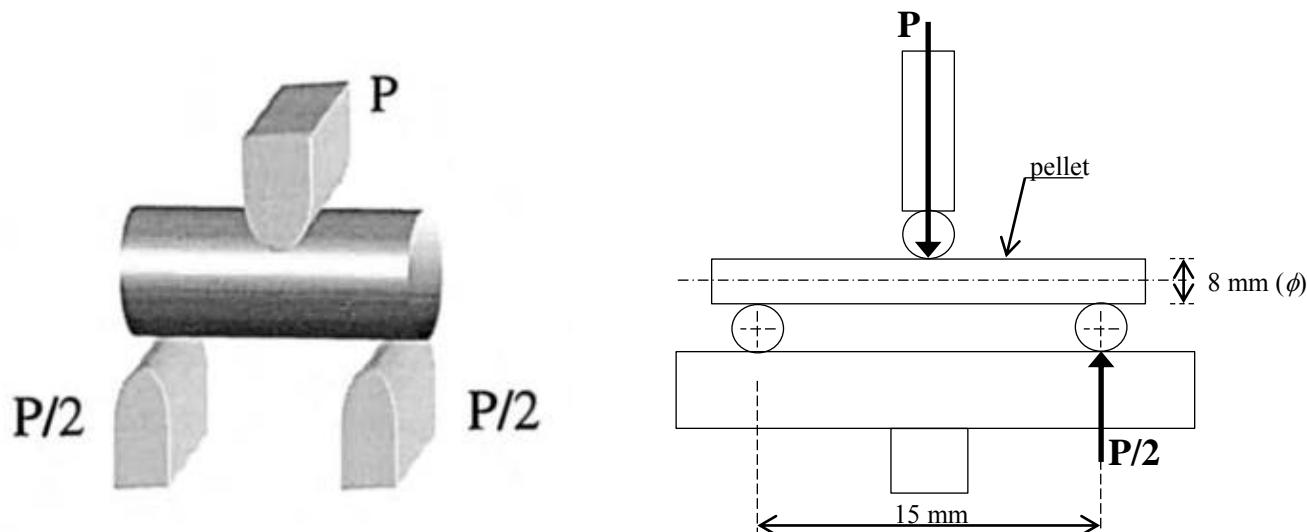


Figure 4. Experimental set-up scheme, pellets testing machine.

A lab made strength tester was utilized in measuring the three-point bend strength data, in order to evaluate the respective rupture strength values. The measurement span between the two lower load points was of 15 mm. It comprised a 50 N cantilever beam load cell from Richmond Industries, with 2.0 mV/V (+/- 10%) sensitivity, a +/- 1mm W1ELA/0-2 inductive displacement sensor connected to a SPIDER8 600Hz data acquisition system, and a Catman 4.5 measurement software, all from HBM.

The correct design of a strength tester is important for getting repeatable and reliable data and is based on a principle of constant rate of loading and on the measurement of the deformation of an elastic cantilever.

The vibration of the testing head and the fluctuations of the loading rate may influence the results though Absi *et al.* (1999) considered that its final effects are small.

For a precise measurement of a material property, many built-in factors affect the precision of the load measurement. In the present work, nonetheless, it is not such a property measurement that is pursued but a comparison between measured values that support comparisons and assess end product quality.

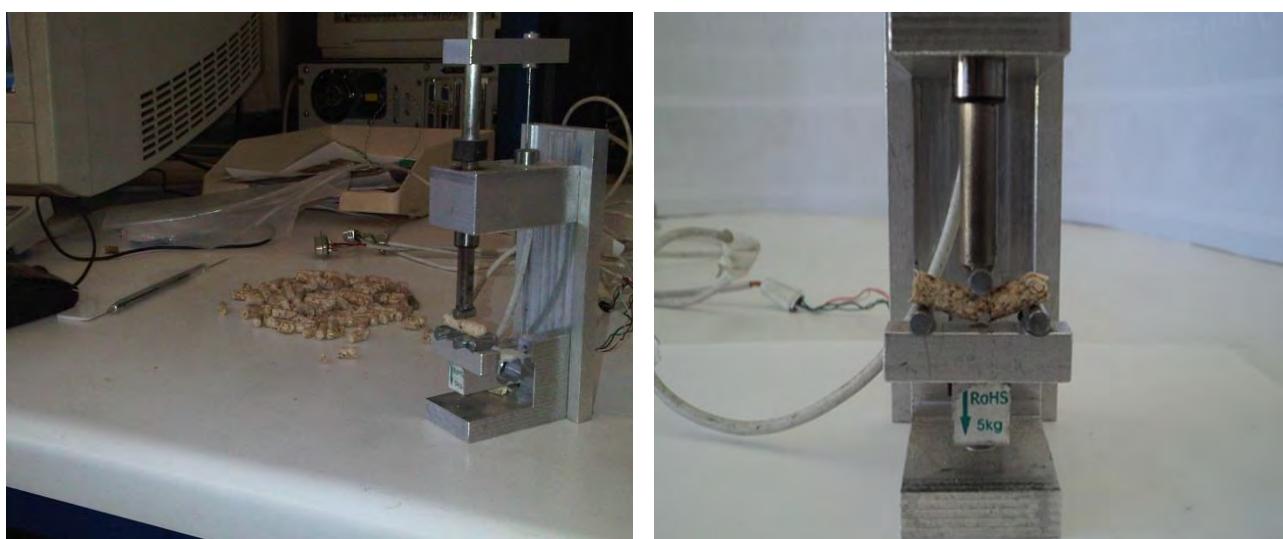


Figure 5. Experimental set-up and photograph of fracture pieces.

Cylindrical shaped pellets were used in these experiments. After the measurement of size, the pellets with length longer than 15 mm, and with no clear curvature or visible flaw, were chosen to be loaded until failure with three-point bending, as shown in Fig. 5.

3.3 Bonding process

The produced pellets have a shiny appearance that, according to Anglés *et al.* (2001), is a result of the lignin coating. This result is an indicator that the applied pressure, the resulting temperature and the existing moisture content during the densification were high enough to cause the softening and flow of the lignin into the pellets surface and the formation of a solid bridge bonding mechanism between the pellets particles that increases its density and its mechanical strength.

For particles orientation studies, longitudinal and transversal sections of pellets were prepared using a surgical blade. Both cross sections were observed under optical microscope and pictures were recorded using a digital camera IDS- UI-1645LE-C-FQ, with CMOS sensors.

In addition to particles orientation, microscope analyses were also used to clarify how particles bond together during the process. These analyses were performed on both kind of pellets, *Acacia dealbata* and *Salix atrocinerea*.

4. EXPERIMENTAL RESULTS

4.1 Manufacturing process

The complete setup was put into work, after being previously set at specified tightening torque, M_t , that produced the initial force F_i (see Fig. 1). Pellets were manufactured at different average material feed rate, for each tightening condition (Tab. 1).

Table 1. Manufacturing pellets material feed rate.

Sample	Initial torque (Nm)	Feed rate (kg/h)
<i>Acacia dealbata</i>	11	120
	13	72
	15	125
	17	131
<i>Salix atrocinerea</i>	11	73
	13	116
	15	81
	17	68

The machine was kept running until an approximate permanent regime was obtained, assessed by measurements of the temperatures of the axis, upper cover and lower tub, Fig.s 6 and 7.

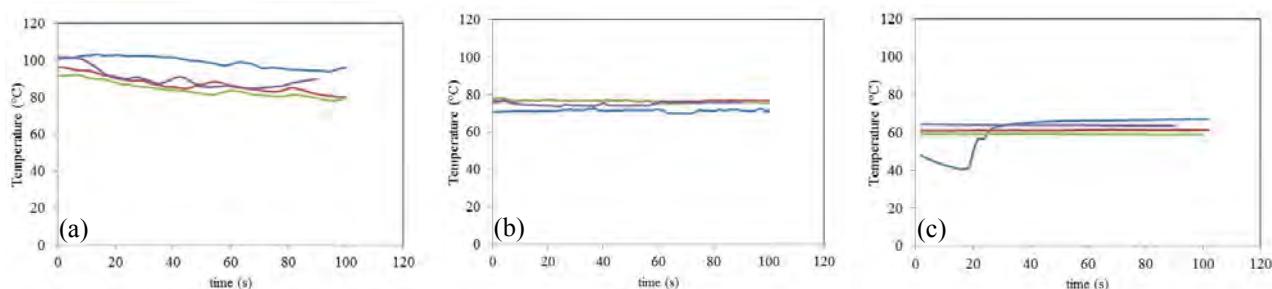


Figure 6. *Acacia* temperature *versus* time for different initial tightening torques: (a) axis, (b) upper cover, (c) lower tub. (violet, 11; green, 13; red, 15; blue 17 Nm).

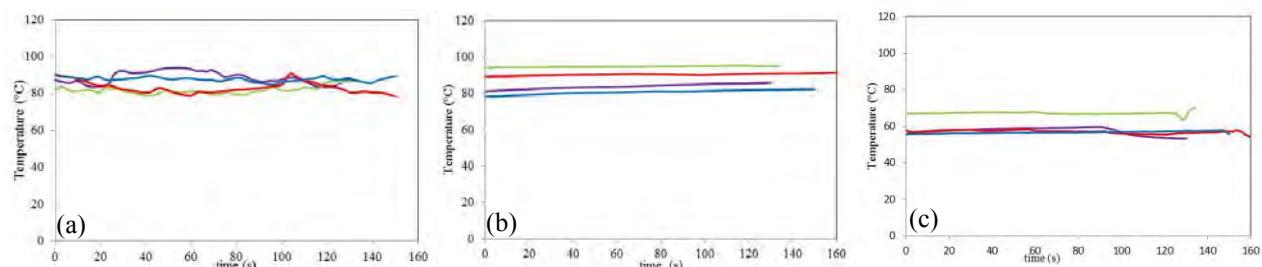


Figure 7. *Salix* temperature *versus* time for different initial tightening torques: (a) axis, (b) upper cover, (c) lower tub. (violet, 11; green, 13; red, 15; blue 17 Nm).

Essays were made with initial torque values of 11, 13, 15 and 17 Nm, for the same materials (*Acacia spp.* and *Salix spp.*). The temperature evolution was observed so that, for periods of approximate permanent regime values, in-time monitored, pellet batches were kept apart every 30 seconds, sequentially. This allowed specific samples of pellets to be identified, correlating the manufactured items properties to those periods of approximate constant temperature.

Table 2. Process temperatures steadiness evaluation.

No	Sample	Average time interval (s)	Torque (Nm)	Manufacturing temperature- axis (°C)	SMD
1	<i>Acacia dealbata</i>	90	11	89.9	4.9
2		100	13	84.1	4.0
3		144	15	85.3	4.6
4		302	17	94.4	4.3
5	<i>Salix atrocinerea</i>	220	11	88.7	3.2
6		130	13	81.9	2.2
7		240	15	83.0	2.9
8		150	17	87.7	1.1

The load forces were registered during the process by means of the load cell and the evolution with time is represented, again for each initial tightening torque of 11, 13, 15 and 17 Nm (Fig. 8).

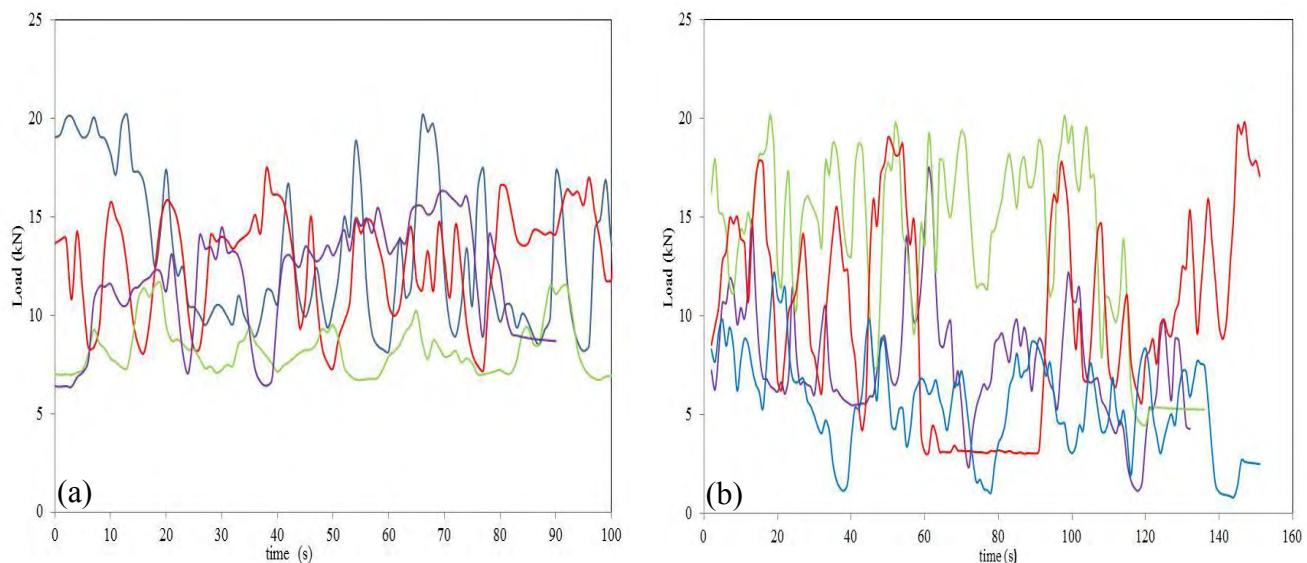


Figure 8. Vertical force values *versus* time for different initial tightening torques: (a) *Acacia*, (b) *Salix*. (violet, 11; green, 13; red, 15; blue 17 Nm).

The behaviour of the system, reflected by the evolution of the measured forces, points out that the pelletization process is significantly intermittent, made of sequences of high values followed by sudden drops of the effort supported by the plate/ load cell system. Some of the lower values reach, inclusively, lesser values than those resulting from the initial applied tightening torque.

4.2 Characterization of *Acacia dealbata* and *Salix atrocinerea* pellets

4.2.1 Physical characterization



Figure 9. *Acacia dealbata*.



Figure 10. *Salix atrocinerea*.

In order to evaluate the characteristics of experimental pellets some tests were carried out. According to ÖNORM M 7135, the dimensions of twenty pellets per sample were measured using a digital caliper with 0.1 mm precision. The corresponding mass also determined using a precision lab scale, Precisa 6200. The particle density of the pellets was determined using the stereometric method (the ratio between pellet weight and volume) calculating the volumes considered as cylinders (Rabier *et al.*, 2006).

The moisture content of pellets was determined according to FprEN 14774-2:2009: pellet samples were dried in a Venticell 50L oven at $105 \pm 2^\circ\text{C}$, until constant mass. The moisture content (*MC*), dry basis, was determined by Eq. (8):

$$MC_{db} (\%) = \frac{m_w - m_d}{m_w} \times 100 \quad (8)$$

where m_w represents the initial wet mass and m_d the dry mass.

Tests of mechanical durability were made for three samples of each type of pellets, using a tumbling device defined by ASAE S 269.4. A 500 g sample was manually sieved with a 3.35 mm ASTM sieve and then tumbled for 500 rotations during 10 minutes (Temmernan *et al.*, 2006). The sample was again sieved and the pellets remaining in the sieve were weighed. The mechanical durability was determined according to Eq. (9):

$$\text{Durability (\%)} = \frac{\text{Mass of pellets after trumblng}}{\text{Mass of pellets before trumblng}} \times 100 \quad (9)$$

The physical characteristics of the pellets manufactured with *Acacia dealbata*, used in the experimental tests, are shown in Tab. 3.

Table 3. Pellets made of *Acacia dealbata*, physical characteristics.

Sample	Property	Initial torque (Nm)			
		11	13	15	17
<i>Acacia dealbata</i>	Diameter (mm)	6.1	6.0	6.2	6.1
	Length (mm)	18.5	20.3	18.7	22.5
	Weight (g)	0.5	0.6	0.5	0.7
	Volume (cm ³)	0.547	0.577	0.557	0.661
	Particle density (kg/m ³)	959.1	1045.5	967.8	1049.5
	Fine content (%)	25.4	38.5	13.8	13.8
	Moisture (%, wb)	7.4	7.1	7.2	6.6
	Durability (%)	73.2	70.6	81.5	89.4
	Water resistance (%)	101.9	31.7	83.9	100.2

Table 4 shows the physical characteristics of the pellets manufactured with *Salix atrocinerea*.

Table 4. Pellets made of *Salix atrocinerea*, physical characteristics.

Sample	Property	Initial torque (Nm)			
		11	13	15	17
<i>Salix atrocinerea</i>	Diameter (mm)	6.0	6.0	5.9	6.0
	Length (mm)	21.1	21.1	18.2	20.4
	Weight (g)	0.6	0.6	0.5	0.6
	Volume (cm ³)	0.600	0.595	0.504	0.570
	Particle density (kg/m ³)	1057.7	1003.7	1063.6	1068.2
	Fine content (%)	6.9	1.2	3.0	3.6
	Moisture (%, wb)	5.6	5.3	5.4	5.6
	Durability (%)	80.3	92.5	85.7	86.2
	Water resistance (%)	73.3	53.8	74.6	50.5

4.2.2 Chemical characterization

The chemical composition includes some primary components, such as cellulose, hemicellulose, lignin, proteins, hydrocarbons and starch. Other compounds that can be found in wooden biomass are lipids, simple sugars and ash, among others. Water, evaluated by the moisture content, is a critical parameter in pelletization. Table 5 contains some of those properties, taken from (Yáñez *et al.*, 2009; Stolarski *et al.*, 2013).

The content of the main components appears similar in the two specimens. The component in major fraction is cellulose, followed by similar fractions of the hemicellulose and acid insoluble lignin. The amount of lignin is in accordance with the range values of $20 \pm 4\%$ (Lange, Decina and Crestini, 2013) found in literature for hardwoods. Salmen (1984) and Irvine (1984) have studied the softening of hemicellulose extracted from wood and have shown that the glass transition temperature depends largely on moisture content and, under water saturated conditions, below room temperature.

The glass transition temperature of lignin also varies considerably, depending on wood species and the moisture content and ranges between 50 and above 100°C (Stelte *et al.*, 2011b).

Table 5. Average chemical composition of Wattle and Willow species (ODM basis).

	unit	<i>Acacia spp.</i> ⁽¹⁾	<i>Salix spp.</i> ⁽²⁾
cellulose	(%, m/m)	42.4	44.1
hemicellulose	(%, m/m)	20.6	21.8
lignin ⁽³⁾	(%, m/m)	19.3	20.4
protein	(%, m/m)	1.60	n.a.
ash	(%, m/m)	0.50	2.10
higher heating value	(kJ/kg)	20088 ⁽⁴⁾	19330

⁽¹⁾Yáñez *et al.*, 2009; ⁽²⁾Stolarski *et al.*, 2013; ⁽³⁾Acid-insoluble lignin; ⁽⁴⁾Godí *et al.*, 2013.

4.3 Three-point bend test results

During the test, all of the pellets were broken into two parts at the center load point, as illustrated in Fig. 4. The results of the strength tests on extruded samples are presented in Table 6, where F_r stands for rupture force.

Table 6. Results of the rupture forces for the three-point bend tests, indexed to the forces acting during manufacturing, ranging from F_{min} , Eq. (5), to F_{max} , Eq. (7), and comparison with homologous durability indexes.

Sample	F_{max} (kN)	F_{min} (kN)	F_r (N)	Durability ⁽¹⁾ (%)
			Mean value	SMD
<i>Acacia dealbata</i>	15.4	6.4	20.0	9.3
	11.5	7.0	19.0	14.1
	17.5	7.6	19.0	11.2
	19.3	8.5	12.8	8.8
<i>Salix atrocinerea</i>	15.0	3.0	3.9	2.9
	19.7	7.4	9.7	8.5
	19.6	7.6	6.1	4.3
	10.2	2.5	5.4	4.2

⁽¹⁾ from Tab.s 3 and 4

4.4 Lignin diffusion analysis

As can be observed, Fig. 11 shows non perfect longitudinal particles alignment and though particles main orientation is on extrusion direction it is clear that many particles bended with a concave region pointing on this direction too (head arrows). Moreover, particles interface are characterized by a homogeneous bonding suggesting a partial softening of particles surface, increasing the interface strength.

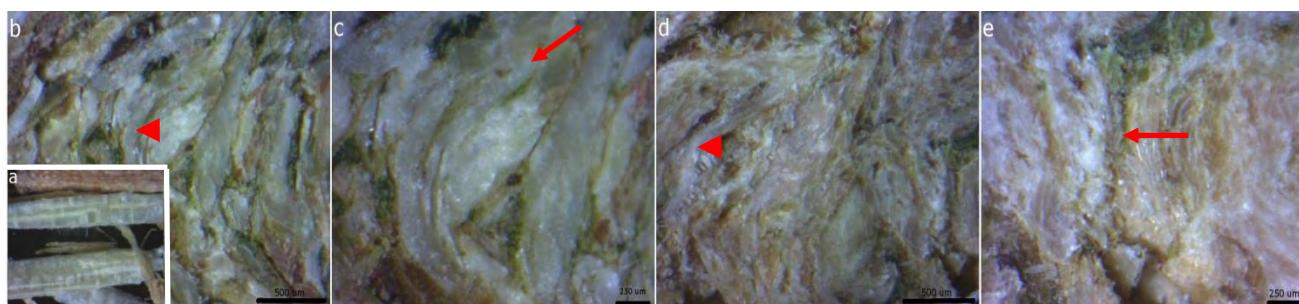


Figure 11. Wood particles and longitudinal cross sections of pellets. Image (a) presents wood particles before pelletization. Images (b) and (c) show longitudinal cross sections of *Acacia* while images (d) and (e), corresponding to the longitudinal cross section of *Salix* pellets, show bended and oriented particles.

Although the interface was not chemically characterized, images show an almost perfect bonding among particles (red arrows). Those observations indicate a partial fusion of polymeric material surface, as a possible result of temperature increase during pelletization process due to the applied pressure and to particles friction against the die. As mentioned before, this process causes a temperature raise to around 100°C. Images from transverse section cuts do not exhibit a pattern sequence (images not presented).

5. RESULTS DISCUSSION

5.1. Influence of the initial tightening torque

The obtained results show few differences between the physical properties of *Acacia dealbata* and *Salix atrocinerea* pellets. Mass and particle densities are slightly higher for *Salix atrocinerea* pellets. *Acacia dealbata* presented higher moisture content values (see Tab.s 3 and 4), below the established by EN 14961-2 (10%). The mechanical durability achieved with both species was also below the minimum established for the same standard (97.5%).

5.1.1 Three-point bend test analysis

Failure strength data of these biomass materials scatter in rather large ranges and do not follow a normal distribution, unlike ductile materials, where plastic deformation takes place before failure. This friable behavior is the main factor affecting the results.

The high mean standard deviation values are a result of significant variations in the compression force during manufacturing (Fig. 8) with reflections in compression ratios conditioning final resistance values.

The homogeneity of the mixture also influences the production process. To discard the temperature influence, average constant periods were identified (Fig. 6 and Tab. 2) so that pellets manufactured during those periods of time were chosen to be tested. It is noted, however, that for the case of willow species the best results occurred for an initial tightening torque of 13 Nm; this behavior corresponds to greater compression forces, both in value and in acting time.

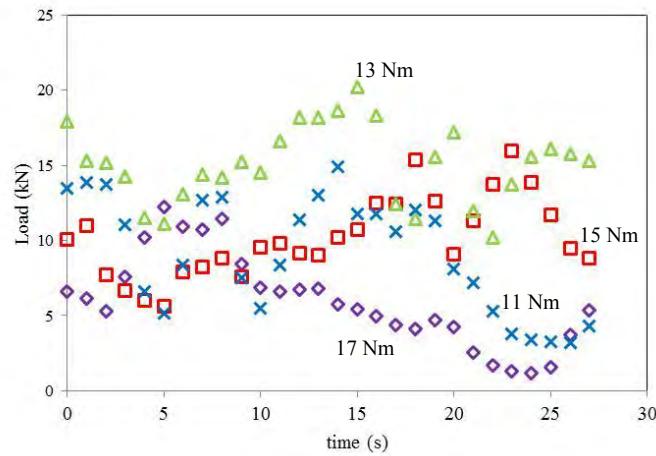


Figure 12. Compression forces (F_c) obtained during manufacturing of *Salix atrocinerea* pellets at constant temperature.

Pellets are made with forces ranging from F_{\min} , Eq. (4), to F_{\max} , Eq. (6), i.e., from the maximum resistance offered by the amount of biomass prior to being extruded, dependent of the particular and momentary physical characteristics, to a minimum value that corresponds to a brief absence of material inside the tub or at the plate/rolls contact surface (Fig. 13); also, transient situations, when biomass material gets stuck to the sides of the hopper, produce that result.

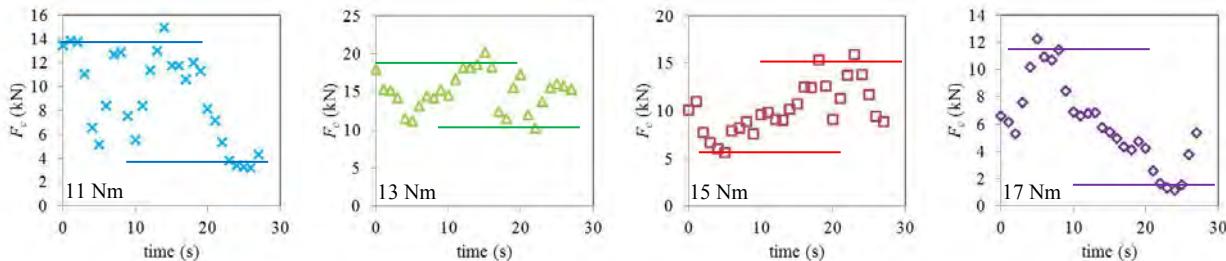


Figure 13. Range of maximum and minimum compression forces during *Salix* pellets manufacturing.

Therefore, for different initial tightening torques applied, there are, in each sample, similar values of rupture strength and quality indexes. The measured specimens that were chosen to create one sample were obtained from the same batch of production batch. These were made of the same material, with the same production procedure and have experienced the same stress conditions as individual pellets.

It can be observed (Tab. 7) that for the *Salix atrocinerea* pellets, the higher rupture force, F_r , is registered for the 13 Nm tightening torque which, in turn, corresponds to the higher average compression force, F_{avg} . In the case of *Salix atrocinerea*, a connection between the average compression force, the rupture force and the durability index was identified based on the experimental results.

Table 7. Results of rupture forces and standard mean deviation for the three-point bend tests.

Sample		F_{avg} (kN)	σ (-)	F_r (N)	σ (-)	Durability ⁽¹⁾ (%)
<i>Acacia dealbata</i>	11	11.8	2.8	20.0	9.3	73.2
	13	8.3	1.3	19.2	14.0	70.6
	15	12.3	0.3	19.0	14.1	81.5
	17	13.8	3.6	12.8	5.4	89.4
<i>Salix atrocinerea</i>	11	9.1	3.7	3.8	2.7	80.3
	13	15.1	2.5	9.9	5.5	92.5
	15	10.2	2.6	5.8	3.5	85.7
	17	6.0	3.1	2.9	1.9	86.2

The pellets production that took place with the higher average forces, along with small mean standard deviations, gave rise to higher tensile strengths and thus higher durability indexes. In the case of *Acacia dealbata* this conclusion is not so obvious because the values of the average compressive forces observed during the manufacturing have very close values, and this observed proximity of the values does not allow identifying clearly the same correlation patterns that are present in the former case analyzed for *Salix atrocinerea*.

5.2 Influence of lignin dispersion

As expected, a physical bonding among particles is an important factor to increase of pellets bending strength. Nonetheless, mechanical behavior is improved if and when a chemical interaction is produced at the particles interface. Since temperatures rise during pelletization, apparently a softening of the lignin occurred easing its diffusion to the interstices of the adjacent particles, the whole bonding process resulting in an increase of the bending strength. These results are in accordance with those obtained by Stelte *et al.* (2011b) for beech pellets processed at the temperature of 100°C. Nonetheless, a direct correlation between the observed pellets cross-sections, the level of lignin dispersion or diffusion near the outer surface regions and the temperatures attained during the pelletization process could not be established by lack of adequate analytical technology.

6. CONCLUSIONS

All specimens exhibited a single fracture mode due to brittle fracture, a synonymous of a constant tensile stress distribution. Higher tightening forces have resulted on higher temperatures and higher values of the average forces. The tests made have shown a correlation between those conditions and the increased rupture forces. There will be an inflection on this trend but, for the present set of data, such point was not detected. Further work will be made in this direction. Lignin softening took place, both with *Acacia dealbata* and *Salix atrocinerea* species, its effects were observed recognizing the lignin dispersion between the interstices of the adjacent particles and, as expected, this effect has resulted in a mechanical strength increase.

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8. REFERENCES

- Absi, J., Fournier, P., Glandus, J., 1999. "Influence of experimental parameters on the estimated value of Weibull's modulus". *J. Mater. Sci.*, 34(6), pp. 1219–1227.
- Anglés, M.N., Ferrando, F., Farriol, X., Salvadó, J., 2001. "Suitability of steam exploded residual softwood for the production of binderless panels. Effect of the pretreatment severity and lignin addition". *Biomass Bioenergy* 21 (3),

- pp. 211–224. Campos, J., Rocha, M. and Tavares, M., 2002. “Controlo de acácas com fitocidas nas dunas do litoral”. *Silva Lusitana* 10, pp. 201–206.
- Dhamodaran, A. and Afzal, T., 2012, “Compression and Springback Properties of Hardwood and Softwood pellets”, *BioResources* 7(3), pp. 4362–4376.
- Direcção-Geral dos Recursos Florestais, 2006. “Estratégia Nacional para as Florestas”, DGRF, Lisboa.
- Elorza, M., Sánchez, D. and Vespertinas, E., 2004. “Atlas de plantas alóctonas invasoras en España”. Ministerio de Medio Ambiente, Madrid, pp. 44–45.
- Faaij, A., 2006. “Bio-energy in Europe: changing technology choices”. *Energy Policy* 34, pp. 322–342.
- Fernandes, M., 2008. “Recuperação Ecológica de Áreas Invadidas por Acacia dealbata Link no Vale no Rio Gerês: um Trabalho de Sísifo?”. Dissertação de Mestrado em Instrumentos e Técnicas de Apoio ao Desenvolvimento Rural. Universidade de Trás-os-Montes e Alto Douro, Vila Real.
- Godí, B. n., Lamaudière, S., Agneessens, R., Schmit, T., Goffart, J. Stilmant, D., Gerin, P. and Delcarte, J., 2013. “Chemical Composition and Biofuel Potentials of a Wide Diversity of Plant Biomasses”, *Energy Fuels*, 2013, 27 (5), pp. 2588–2598, DOI: 10.1021/ef3019244.
- Hatakeyama, H., Hatakeyama, T., 2010. “Thermal properties of isolated and in situ lignin”. In: Heitner, C., Dimmel, D., Schmidt, J.(Eds.), *Lignin and Lignans: Advances in Chemistry*. CRC Press, New York, pp. 301–316.
- Irvine, G.M., 1984. “The glass transitions of lignin and hemicellulose and their measurement by differential thermal analysis”. *Tappi J.*, 67(5), pp. 118–121.
- Kaliyan, N. and Morey, R., 2009. “Factors affecting strength and durability of densified biomass products”, *Biomass and Bioenergy*, Volume 33, Issue 3, pp. 337–359.
- Khan, A., de Jong, W., Jansens and P. Spliethoff, H., 2009. “Biomass combustion in fluidized bed boilers: Potential problems and remedies”. *Fuel Processing Technology* 90(1), pp. 21–50.
- Kitani, O., Jungbluth, T., Peart, R. and Ramdani, A., 1999. “CIGR Handbook of Agricultural Engineering, Volume V- Energy and Biomass Engineering. The International Commission of Agricultural Engineering, published by the American Society of Agricultural Engineers.
- Lestander, T., Finell, M., Samuelsson, R., Arshadi, M. and Thyrel, M., 2012. “Industrial scale biofuel pellet production from blends of unbarked softwood and hardwood stems— the effects of raw material composition and moisture content on pellet quality”. *Fuel Processing Technology* 95, pp. 73–77.
- MADRP, Grupo de Trabalho do MADRP de Energias Alternativas, 2005. “Biomassa e Energias Renováveis na Agricultura, Pescas e Florestas (Ponto da Situação, Junho de 2005)”. Ministério da Agricultura, Desenvolvimento Rural e Pescas, Lisboa.
- Marchante, E., Kjoller, A., Struwe, S. and Freitas, H., 2008. “Short- and long-term impacts of *Acacia longifolia* invasion on the belowground processes of a Mediterranean coastal dune ecosystem”. *Applied Soil Ecology* 40, pp. 210–217.
- Mediavilla, I., Fernández, M. and Esteban, L., 2009. “Optimization of pelletisation and combustion in a boiler of 17.5 kWth for vine shoots and industrial cork residue”. *Fuel Process. Technol.* 90, pp. 621–628.
- Richardson, D. and Rejmánek, M., 2011, “Trees and shrubs as invasive alien species – a global review”. *Divers. Distrib.* 17, pp. 788–809.
- Salmen L., 1984. “Viscoelastic properties of insitu lignin under water-saturated conditions”, *J Mater Science*, 19, pp. 3090–3096.
- Sanadi, A.R., Caulfield, D., 2008. “Thermoplastic polyolefins as formaldehyde free binders in highly filled lignocellulosic panel boards: using glycerine as a processing aid in Kenaf fiber polypropylene boards”. *Mater. Res.* 11 (4), pp. 487–492.
- Santos, A., Simões, R., Pereira, H. and Anjos, O., 2004. “Alternative species for the forest industry as forms of diversify the landscape”. II Simposio Iberoamericano de Gestión y Economía Forestal. Barcelona, Espanha.
- Stelte, W., Clemons, C., Holm, J., Ahrenfeldt, J., Henriksen, U. and Sanadi, A., 2011. “Thermal transitions of the amorphous polymers in wheat straw”. *Industrial Crops and Products*, Volume 34, Issue 1, July 2011, pp. 1053–1056.
- Stelte W., Holm, J.K., Sanadi, A. R., Barsberg, S., Ahrenfeldt, J. and Henriksen, U.B., 2011b. “A study of bonding and failure mechanisms in fuel pellets from different biomass resources. *Biomass and bioenergy*, 35, pp. 910–918.
- Stolarski, M., Szczukowski, S., Tworkowski, J. and Kłasa, A., 2013. “Yield, energy parameters and chemical composition of short-rotation willow biomass”. *Industrial Crops and Products*, Volume 46, pp. 60–65. (<http://dx.doi.org/10.1016/j.indcrop.2013.01.012>)
- Wilson, J., Gairifo, C., Gibson, M., Arianoutsou, M., Bakar, B., Baret, S., Grapow, L., DiTomaso, J., Dufour-Dror, J., Kueffer, C., Kull, C., Hoffmann, J., Impson, F., Loope, L., Marchante, E., Marchante, H., Moore, J., Murphy, D., Tassin, J., Witt, A., Zenni, R. and Richardson, D., 2011. “Risk assessment, eradication, and biological control: global efforts to limit Australian acacia invasions”. *Divers. Distrib.* 17, pp. 1030–1046.
- Li, Y., Zhang, J., Chang, L., Wu, D., Fang, Z., and Shi, Y., 2000. “Measurement and statistics of single pellet mechanical strength of differently shaped catalysts”. *Powder Technology*, 113, pp. 176–184.

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