Evaluation of the papermaking potential of *Ailanthus altissima*

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Abstract

In this work, *Ailanthus altissima* (tree-of-heaven) wood was analyzed for its chemical, morphological and papermaking properties. The *A. altissima* wood was cooked under kraft conditions using different active alkali charges and then handsheets were produced with the pulps having a kappa number of 16. Based on structural, strength and optical data it was found that the kraft pulp of *A. altissima* is not suitable to be used alone for the production of printing and writing papers. Notwithstanding, the handsheets exhibit a favorable value of brightness in comparison to those produced from *Eucalyptus globulus* pulp (with similar kappa number). Therefore, the wood of *A. altissima* seems to have a good potential to be used as a partial substitute of the main raw material of the Portuguese pulp industry. In fact, the results showed that when beaten *E. globulus* and *A. altissima* pulps were mixed (50:50 w/w), the papermaking properties were comparable to those of beaten eucalypts kraft pulps. Therefore, the use of *A. altissima* wood seems promising for the production of uncoated wood-free papers, which has advantages both from an economical and environmental perspective.

Keywords: *Ailanthus altissima*, chemical composition, kraft cooking, papermaking properties

1. Introduction

Ailanthus altissima (Mill.) Swingle (tree-of-heaven) is a member of the Simaroubaceae family. It is a fast growing dioecious species rapidly reaching 15 m of height in about 25 years depending upon the growth conditions. The species is native of the northern and central mainland of China, Taiwan and Japan, where trees can stand for 50 years or more, reaching up to 30 m of height and 1 m of diameter at breast height (Kowarik and Säumel, 2007; Little, 1979). It has spread from oriental countries throughout Europe and USA where it behaves like an invasive plant threatening ecosystems (Fryer, 2010). This is due mainly to its ability to propagate by sexual and asexual means, and also to the allelopathic chemicals it produces, hence inhibiting the growth of neighbor plants (De Feo et al., 2003; Motard et al., 2011). In Portugal this species can be found all over the country being used as an ornamental tree in gardens, parks and streets (Marchante et al., 2008). In addition, this tree has become a pest not only by outcompeting native vegetation, but by causing damage to roadways, sidewalks, sewer structures, and orchards, due to its extensive root system. The plant reproduces very easily since a single female tree can produce as many as 325,000 seeds per year, which are easily dispersed by the wind (Dirr, 2009). It prefers rich and moist soils but tolerates also poor and dry soils. Besides, tree-of-heaven tolerates air pollution and may be able to sequester some pollutants. For this reason, it has been widely planted in urban areas worldwide to reduce the environmental pollution (Ding et al., 2006a; Ding et al., 2006b; Kundu and Laskar, 2010).

Eucalypt is one of the most valuable resources for papermaking worldwide, Portugal being the European largest producer of uncoated wood-free papers based on *Eucalyptus globulus* kraft pulps, which are recognized for the high multi-purpose printing quality.

However, due to the extensive use, scarceness, high prices in the market and distribution problems associated to eucalypt pulps, it is advantageous to find alternative wood resources. In this context, *A. altissima* seems to be an interesting tree to be tested. Recently, *A. altissima* has been delignified using alkaline glycerol (Kuçuk, 2005; Kuçuk and Dermibas, 1993). A pulp yield of 54.5%, a delignification degree of 78.1% (after 9 hours of reaction at 478 K) and a remaining cellulose content of 86% were obtained when NaOH was used as catalyst in the process. In this paper, the use of *A. altissima* wood for kraft pulping aiming to obtain pulps with valuable properties for papermaking is reported by the first time.

2. Material and methods

Since no plantations of *A. altissima* are available, wood of this species was collected at breast height from a tree in the Botanical Garden of the University of Coimbra. The tree was about 20 years old, which corresponds to half of its life span. The trunk had a 20 cm diameter, lower than that of older trees. The chips were air dried, screened (SCAN-CM 40) and ground (TAPPI Standard T 264) for their chemical composition analysis (lignin, carbohydrates, uronic acids, ash and extractives). Wood samples were macerated for the determination of the fibers morphological parameters.

A rotatory digester was used to cook 200 g (o.d.) of chips at 160 ºC, with a heating rate of 1 ºC/min. The liquid to wood ratio was 3.5:1 and the time at maximum temperature was 60 min. Four different cooks with active alkali charges of 14%, 16%, 18% and 20% (as Na_2O) were performed, always with a sulfidity of 28%. After cooking, the total and screened yield was determined and the pulp with a kappa number of 16 was analyzed for its chemical composition (lignin, carbohydrates and extractives), intrinsic viscosity and fiber biometry.

For both the wood and pulp samples, the content in Klason and acid-soluble lignin was determined using T 222 om and T UM 250 TAPPI Standard methods, respectively. Carbohydrates were determined by hydrolysis of the extracted wood or pulp samples firstly with 72% H_2SO_4 during 2 h at room temperature and then with 4% H_2SO_4 during 4 h at boiling temperature, followed by separation of the sugars by high pressure liquid chromatography (HPLC), using a Knauer HPLC instrument with refractive index detector and a HPX-87P column from Bio-Rad®. The eluent used was ultrapure water and the operating conditions were 0.6 ml/min and 80 ºC. The extractives (ethanol/toluene) and ash amount were obtained following the T 204 cm and the T 211 om TAPPI Standards, respectively. Uronic acids in woods were determined following a published procedure (Scott, 1979). A Jasco V-550 spectrophotometer was used for the absorbance measurements. Pulps viscosity and kappa number were determined according to SCAN-CM 15 and TAPPI Standard T 236 cm, respectively.

The biometry of the fibers (mean fiber length, fiber length distribution, fines content and fibers coarseness) were evaluated with the OpTest HiRes Fiber Quality Analyzer (FQA), using very dilute sample suspensions. Average values for the fiber width and fiber wall thickness were evaluated by image analysis, by taking measurements in individual fibers, using a CCD camera coupled to a light microscope

A sample of *E. globulus* unbleached kraft pulp with a kappa number of 16 was obtained from a Portuguese mill for comparison purposes, and its chemical composition was determined as described above.

UV-vis diffuse reflectance spectra of the pulps were recorded on a Jasco V-560 spectrophotometer with a Jasco ISV-469 integrating sphere using $BaSO_4$ standard as background reference. The acquisition was done in the range of 200-800 nm with a scanning speed of 100 nm / min and a bandwidth of 5 nm. The reflectance spectra were converted into k/s spectra using the Kubelka-Munk equation (k stands for the specific absorption coefficient and s is the specific scattering coefficient). Pellets for analysis were prepared by pressing approximately 300 mg of ground pulp for 2 min at 40 MPa.

Handsheets with a basis weight of 65 g/m^2 were prepared accordingly to the TAPPI Standard T 205, using both the *A. altissima* and *E. globulus* kraft pulps with 16 kappa number. Afterward, structural (thickness, bulk, air resistance (Gurley), and roughness (Bendtsen), optical (R457 C brightness, light scattering and opacity), and mechanical properties (tensile index, tensile stiffness, elongation, Scott bond, burst index, tear index) were determined following the corresponding TAPPI Standards. Pulps drainability was also determined. Handsheets with PFI beaten pulps (having a SR close to 30) were also produced and the aforementioned properties were evaluated.

3. Results and Discussion

3.1 Chemical Composition and Fibers Biometry of Ailanthus altissima Wood

The amounts of extractives, lignin (Klason, acid-soluble and total lignin), cellulose, hemicelluloses and ash determined for *A. altissima* wood are presented in Table 1. Typical data for *E. globulus* are also presented for comparison and are in reasonable agreement with others reported in the literature for this wood species (Pinto et al., 2005).

The values for Klason lignin and total lignin are slightly lower than those reported by Khattak and Ghazi (2001) and Kuçuk (2005): 22.5 and 26.5%, respectively. On the other hand, the extractives content (≈ 2%), is similar to that found for this wood by Kuçuk (2005). As for the carbohydrate composition of *A. altissima* wood, it has not yet been reported in the literature and thus no comparison can be made. When compared to *E. globulus* wood, the *A. altissima* wood presents a smaller amount of total lignin, mostly due to the lower value of the acid-soluble lignin, but a larger amount of ash. Regarding polysaccharides, higher content of cellulose and galactan and lower content of xylan and glucomannan are found in *A. altissima* wood. Uronic acids and arabinnan contents are similar for both woods.

Please, Insert table 1

Concerning the fiber morphology analysis for the *A. altissima* wood the average fiber length determined is 0.74 mm for L_l (length weighted) and 0.54 mm for L_n (number weighted), the fiber width and the fiber wall thickness are approximately 20 μm and 4 μm, respectively. The length and fiber width values are not far from those previously found for the same species by Khattak and Ghazi (2001): 0.89 ± 0.2 mm, 21.05 ± 1.88 μm and 2.43 ± 0.3 μm, respectively for L_l fiber length, width and wall thickness.

3.2 Pulp Characterization

The results of the different cooks of *A. altissima* wood chips are presented in Table 2. As expected, it was found that the increase of the active alkali charge (from 14 to 20% as Na_2O basis) leads to a decrease of the pulps kappa number, ranging from 21.6 to

12.6. The pulp yields were in the range of 52 to 55% being comparable to those of *E. globulus* kraft cookings (Carvalho et al., 2003).

Please, Insert table 2

The chemical analyses of the *A. altissima* pulp selected for the subsequent papermaking tests (Table 3) show that this pulp presents higher cellulose and lower xylan content than the *E. globulus* pulp for the same kappa number. The residual lignin content and the pulp intrinsic viscosity are similar for both pulps. Therefore the selectivity of both cooking processes expressed in terms of pulp viscosity/residual lignin is comparable.

Please, Insert table 3

As for the fiber morphology, it is clear from Table 4 that the *A. altissima* kraft pulp fibers are longer and larger than those of *E. globulus* and, in agreement, have a larger wall thickness, a higher coarseness, a higher Runkel index and a smaller number of fibers per gram. They also exhibit a considerably higher amount of fines. These differences will be reflected in the beatability and the papermaking properties, as discussed next.

Please, Insert table 4

3.3 Papermaking properties

The properties of the handsheets prepared with the *A. altissima* and industrial *E. globulus* kraft pulps with kappa 16 are presented in Table 5.

As can be seen, the drainability of the unbeaten *A. altissima* pulp is higher than that of the *E. globulus* pulp, in agreement with the higher values of the fibers coarseness, width and wall thickness, which reduce the fibers flexibility and colapsability and, therefore, increases the porosity of the fiber mat. The results also reveal that the effect of the smaller conformability of the *A. altissima* fibers in the drainability is higher than the effect of the higher fines content of the pulp. The values of the handsheets bulk, air resistance and roughness, for the unbeaten pulps, are also a consequence of the fibers morphology and of the resultant more open structure of the fibers matrix.

Regarding the strength properties, it is clear that the unbeaten *A. altissima* fibers have a reduced bonding ability, in accordance with the aforementioned smaller conformability. In addition, due to the higher rigidity of the fibers, the beating time (in terms of PFI rev.), necessary to reach a Schopper-Riegler degree (SR) close to that of the eucalyptus pulp corresponding to a tensile index of approximately 70 Nm/g, is much higher. Besides, the smaller xylan content of the *A. altissima* pulp and hence the inferior swelling ability of the fibers is also detrimental for its beatability.

Please, Insert table 5

It should be mentioned the significantly higher reflectance at 457 nm of the *A. altissima* kraft pulp, compared to that of the *E. globulus* pulp, which is an important feature for the subsequent bleaching treatment and paper optical properties. Further confirmation comes from the UV-vis electronic reflectance spectra of Figure 1. In fact, the *k*/*s* spectrum of *A. altissima* pulp shows much lower absorption in all the visible range, including the wavelength of 457 nm, when compared to that of *E. globulus* pulp. Besides, in the UV region, *A. altissima* presents an absorption maximum at 335 nm while *E. globulus* shows the absorption maxima at 340 and 370 nm (the latter is very close to that of 365 nm reported for *Eucalyptus* kraft lignin (Loureiro et al., 2011). Thus, since the bands with maxima at $\lambda \geq 300$ nm are due to the presence of chromophore groups in lignocellulosic pulps (Loureiro et al., 2011) a lower amount is anticipated for *A. altissima*, which should be responsible for the higher brightness exhibited by this pulp.

Please, Insert Figure 1

With refining it is expected that the *E. globulus* fibers develop internal fibrillation better than the *A. altissima* fibers, due to the higher amount of hemicelluloses. For the stiff *A. altissima* fibers, external fibrillation and cutting are expected to be the dominant refining effects. As a consequence of these phenomena and of the more extensive beating operation, the predictable reduction of bulk, roughness and air resistance is much more pronounced for the *A. altissima*, and thus the beaten pulp exhibits values close to those of the *E. globulus* beaten pulp (contrary to the unbeaten pulps). Also the strength properties of the *A. altissima* pulp are considerably developed with refining but, even though, the handsheets mechanical resistance remains below the one of the *E. globulus* beaten pulp. This is most probably a result of the weaker internal fibrillation and of the smaller xylan content, which are recognized to play an important role on the fibers flexibility and bonding ability (Laine et al., 1997; Paavilainen, 1994). The exceptions are the tensile stiffness and the handsheets internal resistance (evaluated by the Scott Bond test), probably due on the one hand to the influence of the fibers rigidity and on the other hand to the effect of the external fibrillation. Both light scattering and opacity decrease with refining, as expected, being the variation of the *A. altissima* pulp more noticeable. The values of these two properties are even inferior to those of the *E. globulus* beaten pulp, as well as the value of bulk, confirming that the *A. altissima* handsheets are slightly less porous, probably due to the dominant effects of external fibrillation and cutting, which promote fines formation. These fines play a relevant filling role but their impact on the strength properties is scarce.

In spite of the weak strength properties of the *A. altissima* pulps it was decided to mix them with the *E. globulus* pulps in order to evaluate the papermaking potential of the resulting blend. Therefore, a 50:50 (w/w) mixture of the beaten pulps of both species was prepared. The properties of the corresponding handsheets are also presented in Table 5. As it is clear, the strength properties are almost as good as those of the *E. globulus* beaten pulp handsheets (the internal bond – Scott Bond test – is even considerably better) and a not relevant variation of bulk, opacity and light scattering is found. However, as for brightness, the aimed positive influence of the *A. altissima* beaten fibers was not detected, since the final value is close to the one of the original *E. globulus* beaten pulp. Even so, it is interesting to conclude that the substitution of half of the *E. globulus* beaten pulp furnish by an equal amount of *A. altissima* beaten pulp is not harmful for the papermaking properties which are important for the common end use of the eucalyptus kraft pulps – the production of printing and writing fine papers. Nonetheless, it must be stressed that, as revealed by this study, the refining energy necessary to enhance the papermaking potential of the *A. altissima* stiff fibers is too high, if compared to that of the *E. globulus* pulps for a similar refining degree.

Nevertheless, the results obtained are promising and show that there is room to keep on with this research. On the one hand the use of less refining energy for the *A. altissima* kraft pulps (and, consequently, a smaller degradation of the fibers) should be tested, as well as refining together mixtures of unbeaten *A. altissima* and *E. globulus* kraft pulps, trying simultaneously blends with distinct contents of both pulps. Bleaching performance needs also to be studied, in face of the much better brightness of the *A. altissima* kraft pulp fibers. The smaller content of hemicelluloses of these fibers, and the subsequent detrimental effect on their bonding ability, can also be somehow overcome by adding hydrophilic polymers like starch, during the papermaking process.

4. Conclusions

Ailanthus altissima wood exhibits a lower amount of lignin and xylan and a higher amount of cellulose, when compared to *E. globulus* wood. The corresponding kraft pulps with kappa number of 16 have similar lignin content but *A. altissima* pulp exhibits also a higher cellulose/xylan ratio.

Ailanthus altissima pulp fibers are longer, wider and have also a larger wall thickness than *E. globulus* fibers.

The handsheets prepared with *A. altissima* unbeaten kraft pulp (kappa number of 16), exhibit mechanical properties significantly worse than those of *E. globulus* unbeaten pulp handsheets, namely tensile index, elongation, Scott bond, burst index and tear index. However, the brightness of *A. altissima* is significantly higher, which is an advantage for papermaking.

The strength properties of the *A. altissima* pulp are considerably improved with refining, although they remain below those of the *E. globulus* beaten pulp for similar SR (except the tensile stiffness and the handsheets internal resistance).

When mixtures 50:50 (w/w) of beaten *E. globulus* and *A. altissima* pulps are used for the production of handsheets the results regarding strength properties are similar to those obtained with beaten (750 rev) *E. globulus* pulp alone. As for brightness, the positive influence of the *A. altissima* beaten fibers was somewhat diminished in the mixtures, since the final value is more close to the one of the original *E. globulus* beaten pulp.

The use of *A. altissima* wood can be an alternative to partially reduce the dependence of other species such as *E. globulus* for papermaking. The lower amount of xylan in *A. altissima* pulps and its negative effect in handsheets strength properties can be possibly overcome by adding appropriate amounts of polymers with binding properties like starch, during the papermaking process.

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Table 1. Chemical composition of *A. altissima* and *E. globulus* woods

Components (% w/w, on o.d. wood)	*A. altissima*	*E. globulus*
Extractives (ethanol/toluene)	1.8	1.1
Klason lignin	21.2	20.9
Acid-soluble lignin	2.4	4.8
Total lignin	23.6	25.7
Cellulose	49.7	45.5
Xylan[a]	12.6	14.8
Glucomannan[b]	0.6	1.5
Galactan	4.1	1.5
Arabinnan	0.6	0.5
Uronic acids	6.6	6.2
Ash content	0.9	0.3

[a] The xylan content value presented does not include the methylglucuronic or acetyl groups as substituents

[b] Assumes mannose:glucose ratio of 2:1

Table 2. Pulp yields and kappa numbers from *A. altissima* kraft cooking

Active alkali (as Na_2O) (%)	14	16	18	20
Total yield (%)	55.2	54.9	54.0	52.1
Rejects (%)	0.6	0.6	0.3	0.2
Screened yield (%)	54.6	54.3	53.7	51.9
Kappa number	21.6	16.0	13.9	12.6

Table 3. Properties and chemical composition (% w/w) of the *A. altissima* and industrial *E. globulus* kraft pulps (unbleached and unbeaten pulps with a kappa number of 16)

	A. altissima kraft pulp	*E. globulus* kraft pulp
Viscosity (cm^3/g)	1110	1120
Extractives (ethanol/toluene)	0.3	0.2
Cellulose	77.0	70.9
Xylan	11.8	17.3
Klason lignin	1.4	1.0
Acid-soluble lignin	0.8	1.0
Total lignin	2.2	2.0

Table 4. Morphological properties of the *A. altissima* and industrial *E. globulus* kraft pulp fibers (unbleached and unbeaten pulps with a kappa number of 16)

	A. altissima kraft pulp	*E. globulus* kraft pulp
Fines content (< 0.150 mm) (%)	12.2	7.7
Fiber length (L_n) (mm)	0.67	0.56
Fiber length (L_l) (mm)	0.80	0.66
Fiber coarseness (mg/m)	0.122	0.087
N^{o} fibers ($\times 10^{-6}$) / g	10.3	17.5
Fiber width (μm)	24.2	16
Fiber wall thickness (μm)	4.8	2.5
Runkel index	0.66	0.45

Table 5. Papermaking properties of the unbleached (kappa number of 16) *A. altissima* and *E. globulus* kraft pulp handsheets

	A. altissima (AA) kraft pulp		*E. globulus* (EG) kraft pulp		EG:AA (50:50, w/w)[a]
Beating time (rev. PFI)	0	3000	0	750	
Drainability (SR)	14	31	18	34	
Basis weight (g/m^2)	63.6	64.0	63.8	64.8	65.3
Bulk (cm^3/g)	1.99	1.42	1.65	1.53	1.45
Air resistance (Gurley, s/100 ml)	0.7	9.0	1.8	9.5	8.2
Roughness (smooth side, mL/min)	805	165	367	167	164
R457 C	44.5	44.3	36.2	35.0	36.8
Light scattering (m^2/kg)	36	23	34	28	25
Opacity (%)	95.3	90.1	96.5	95.4	93.2
Tensile index (N.m/g)	23.0	63.5	40.6	69.1	68.4
Tensile stiffness (kN/m)	460	615	490	555	560
Elongation (%)	0.5	2.7	1.9	3.1	3.0
Scott bond (J/m^2)	64	321	128	207	259
Burst index (kPa.m^2/g)	0.68	3.67	1.71	4.28	4.19
Tear index (mN.m^2/g)	2.5	7.4	5.6	9.4	8.6

[a] Mixture of refined *E. globulus* (750 PFI) and *A. altissima* (3000 PFI) kraft pulps

Figure captions

Figure 1. UV-vis *k*/*s* spectra of *A. altissima* (a) and *E. globulus* pulps (b).

Figure

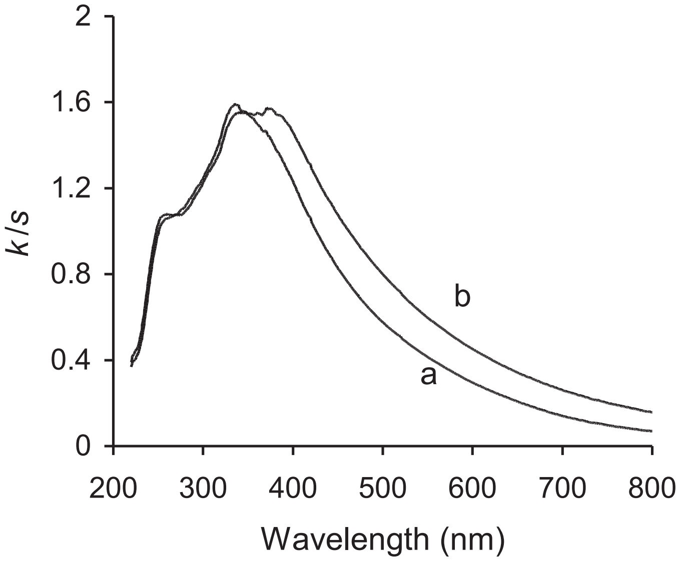

Fig. 1