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INVASIVE *ACACIA* SPECIES' WOOD TO PAPER

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SUMMARY

Acacia species have invasive status in southern Mediterranean European countries with control initiatives generating large biomass waste. An alternative use for *Acacia* wood wastes could add to the pulp and paper industry raw material needs. This work assessed the pulpwood potential of five Acacia species (*A. dealbata, A. longifolia, A. mearnsii, A. melanoxylo*n and *A. saligna*) for bleached kraft pulp and paper production. Response Surface Methodology was used to correlate alkali charge and reaction temperature on pulping yield and kappa number. Optimal pulping conditions were determined for each species. Bleached and refined (2500 Rev) pulp and paper characteristics were also determined. *Eucalyptus globulus* wood was used for comparison. *Acacia* pulp yields varied between 52.5-55% and kappa number between 14.7-18.3 at the optimal conditions. Regarding pulp and paper characteristics, strength properties showed promising results, with tensile and tear index achieving similar results to *E. globulus*. The optical properties showed acceptable original brightness although reducing significantly with beating to 64-76% for *Acacia* species while remaining almost unaltered for *E. globulus* 81%. Overall results showed bleached kraft paper could be a promising end-use for *Acacia* species wood obtained from control actions, which could encourage further initiatives and help mitigate their propagation.

KEY-WORDS: Response surface methodology, kraft pulping, fiber morphology, handsheets properties

INTRODUCTION

Acacia is a genus widely spread as an intentional or invasive populations [1]. These species have a rapid growth rate, high biomass production, robust seed banks, and nitrogen-fixing capacity [2]. They can decrease water availability and increase wildfire frequency and severity and changes in soil nutrient levels [3].

In southern European Mediterranean countries (Portugal, Spain, France, Italy), the most predominant acacias have invasive species status [4]. Control initiatives from local authorities generate significant amounts of biomass waste, mainly used for energy production [4].

Acacia wood is an interesting raw material for pulp and paper production. Some species are already used for this purpose, mainly in Africa, East Asia, South America and Australia[5,6].

Deep knowledge of raw material is required to assess the applicability for pulp and paper production: structural and chemical characteristics, pulping and bleaching are the main variables that influence delignification and their effect on fibers, and final product (pulp and paper) characteristics.

In this work, we studied the pulpwood suitability of five *Acacia* species' wood residues from invasive species control actions, for bleached kraft pulp and paper production (*A. dealbata, A. longifolia, A.*

mearnsii, *A melanoxylon* and *A. saligna*). A central composite design was produced for each species to modulate the correlation between alkali charge and reaction temperature on pulping yield and delignification extension. Optimal pulping points were determined using the models, and the pulp was bleaching through an elemental chlorine-free bleaching sequence. The pulps were refined, and the characteristics of the pulp and paper were assessed. *Eucalyptus globulus* wood was used as a reference.

MATERIALS AND METHODS

Sampling

Wood from *A. longifolia* and *A. melanoxylon* were collected from the fields of Monte da Lua, at Parques de Sintra, Sintra, Portugal. *A. dealbata*, *A. mearnsii* and *A. saligna* were collected from fields of the School of Agriculture, University of Lisbon (ULisboa), at Tapada da Ajuda, Lisboa, Portugal. Wood chips for pulping were obtained through a knife-mill and sieved through a 10x10mm mesh. From this point forward, *A. dealbata*, *A. longifolia*, *A. mearnsii*, *A. melanoxylon* and *A. saligna* will be abbreviated to Ad, Al, Am, and As, respectively. *Eucalyptus globulus* (henceforth Eg) wood chips were provided by the Navigator company and collected at the Setúbal pulp mill, in Portugal.

Kraft pulping

Stainless steel micro digesters (Ca 100mL) under rotation in an oil bath were used to produce kraft pulps. The reaction was achieved using 10 g of wood with a liquid-to-wood ratio of 4:1, 25% sulfidity over 60 minutes under isothermal conditions.

The parmeters studied were active alkali (AA, as Na_2O), ranging from 16% to 24% and temperature (T, ^oC), ranging from 151 ^oC to 179 ^oC). The solid residues were washed, defibrated, further washed and recovered by vacuum filtration. The pulp was dried at 50°C and stored before further tests. A response surface methodology was used to determine the minimum necessary points required to correlate the effects of active alkali (AA) and temperature (T) and the response variables yield (Y) and Kappa number (K). A central composite design was produced with 4 factorial points, 4 axial points and a central point.

Scale-up

To scale-up the process, kraft pulping was performed in a stainless-steel batch reactor (Ca 5L) with fluid recirculation, under the optimized cooking conditions previously established for each *Acacia* species.

E. globulus pulp was produced with the average conditions of AA and T determined for all 5 *Acacia*: AA=17% and T=169 ^oC. The reaction time was determined to obtain the same H factor achieved in the optimized point of the microdigesters assays. The H factors used for the optimum points were 607 for *A. dealbata* (Ad), 846 for *A. longifolia* (Al), 660 for *A. mearnsii* (Am), 1082 for *A. melanoxylon* (Amx), 1173 for *A. saligna* (As), and 846 for *E. globulus* (Eg). After pulping the pulps were thoroughly washed, disintegrated and screened for shives and uncooked material removal.

Kraft pulp characterization

Pulp yield was calculated as the pulp weight obtained from the wood sample, expressed as the over-dry mass percentage. The Kappa number of the pulp was determined according to Tappi Useful Test Method UM 246.

Bleaching

An elemental chlorine-free (ECF) sequence of oxidation with chloride dioxide (D) followed by alkaline extraction (E) (D0-E1-D1-E2-D2) was used to bleach the pulps. The D0 stage was conducted at 45 ^oC, for 30 min, with a chlorine dioxide charge (expressed as active chlorine) corresponding to a Kappa factor of 0.2 (charge % = Kappa number x 0.2). The chlorine dioxide charge (expressed as active chlorine) was 1.3% and 0.6% in D1 and D2. The D1 stage was conducted at 70 ^oC, during 120 min, and the D2 stage was carried out at 70 ^oC, during 180 min, with a consistency of 10%. The bleaching performance was evaluated by measuring the brightness and the intrinsic viscosity (SCAN-CM 15:88).

Papermaking potential

The bleached pulps were beaten at 2500 revolutions in a PFI mill under a refining intensity of 1.77 $N.mm^{-1}$ (as defined in ISO 5264-2). Schopper-Riegler degree (^oSR) was determined according to ISO 5267-1. A Morfi® (LB-01) analyzer developed by Techpap (France) was used to study the morphological properties of the pulp fibers. Handsheets were produced with a basis weight of 60 $g.m^{-2}$ and conditioned according to ISO 5269-1 and ISO 187. The properties measured were: tensile index, tear index, and optical properties (brightness and opacity, according to ISO 2470 and 2471, respectively).

RESULTADOS E DISCUSSÃO

Kraft pulping experimental design and optimization

The pulp yield (g of pulp per 100g of wood) obtained varied between 46-58% and the Kappa number (representing residual lignin and hexenuronic acids) between 6-60. As expected, harsher conditions (higher temperatures and/or chemical charges) lead to lower pulp yield and Kappa number due to higher degradation and solubilization of both lignin and carbohydrate polymers (especially hemicelluloses).

Acacia saligna showed higher Kappa number values and lower pulp yields for almost all experimental runs than the remaining species. Regarding delignification, *Acacia melanoxylon* showed Kappa number varying solely from 9-38, appearing to be easier to delignified at softer conditions within the experiment domain.

Each *Acacia* optimized points of temperature and active alkali were used to produce pulp whose yield and Kappa number were comparable to the theoretical values extracted from the models. The main deviation happened for *Acacia melanoxylon,* with a lower Kappa number than expected.

The optimized results were then used to upscale and produce enough pulp to proceed with the pulp and paper production and characterization. A pulp produced with *Eucalyptus globulus* was used as the standard for comparison reasons. Results of unbleached and bleached pulps of each species are presented in Table 1.

The results suggest that the scale-up procedure lead to higher polysaccharides degradation, higher solubilization and removal of hexenuronic acids since the yields obtained were lower than those expected from the pulps produced in small digesters. *A. longifolia* was the only species that did not reach a kappa number of 17. The pulps bleaching results showed mass losses of 4-5% and almost complete removal of hexenuronic acids. These delignification variables indicate that *Acacia* species might be an interesting raw material for pulping, with yields and delignification patterns comparable to *E. globulus*.

Table 1. Optimum pulping values of temperature (T) and Active Alkali (AA), Pulp yield (Y), Kappa number (K), hexenuronic acids component in Kappa number (HexA), and pulp degree of polymerization (DP), for Response Surface Methodology theoretical and experimental optimal points, and scale-up unbleached and bleached pulps.

	Optimum factors values		Optimum point Scale-up						
			Unbleached				Bleached		
	T (°C)	AA (%)	Y (%)	K	HexA	DP	Y (%)	HexA	DP
Ad	165	17	54	16	3.6	3514	96	0.2	3203
Al	169	16	53	19	4.3	3490	96	0.2	3086
Am	166	18	53	15	3.4	3305	95	0.2	2956
Amx	172	16	52	17	3.8	4215	96	0.2	3690
As	173	16	53	18	4.1	3211	95	0.2	2867
Eg	170	17	51	16	4.1	3242	96	0.1	2835

Papermaking potential

The papermaking potential of *Acacia* was assessed with unrefined, and 2500 revolutions refined bleached pulps.

Fiber properties

The morphology of *Acacia* fibers is shown in Figure 1. *A. melanoxylon* is the only species with lower values than *E. globulus*. However, it has higher amounts of fine elements. *Acacia* fibers belong to the "short" fiber category. *A. mearnsii* and *A. saligna* presented fiber widths similar to *E. globulus*. *A. longifolia* have considerably higher average fiber width. Regarding coarseness, *A. longifolia* has the highest coarseness among the studied species. *A. dealbata and A. saligna* have lower values which indicate that the fiber's walls are thinner than those of *E. globulus*. In the case of *A. longifolia*, these results do not imply low collapsibility because it has a higher width.

Beating the pulps resulted in a slightly decrease in fiber length and an increase in fines, as expected. This happens because of the severance of the fiber extremities upon beating. Width increases because of higher surface fiber microfibrillations and reduction of the fibers' extremities.

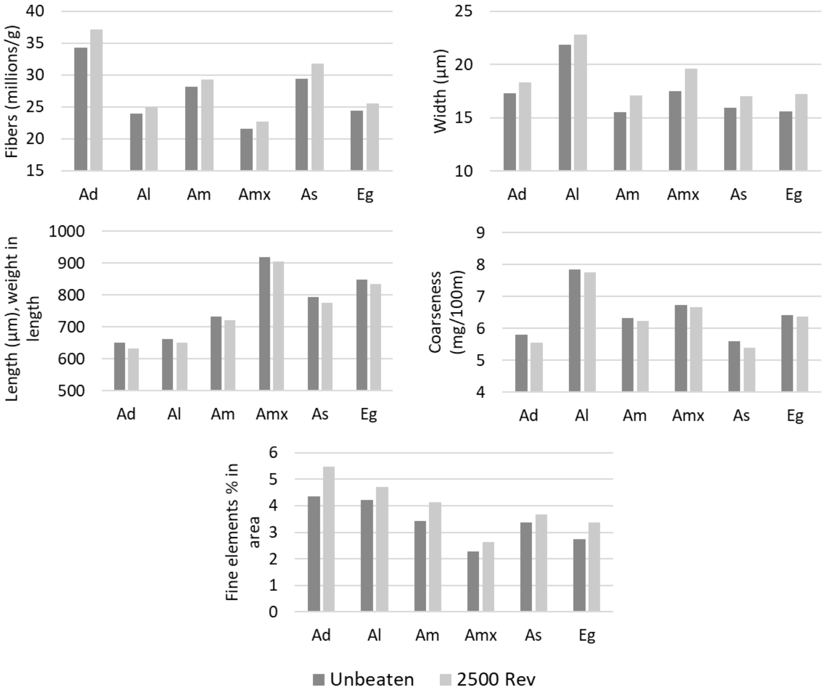

Figure 1. Morphological properties of pulp fibers produced from *A. dealbata* (Ad), *A. longifolia* (Al), *A. mearnsii* (Am), *A. melanoxylon* (Amx), *A. saligna* (As) and comparison to *Eucalyptus globulus* (Eg)

Pulp and Handsheet properties

Figure 2 shows the handsheet physical, strength, and optical properties of unbeaten and beaten pulps of the studied species.

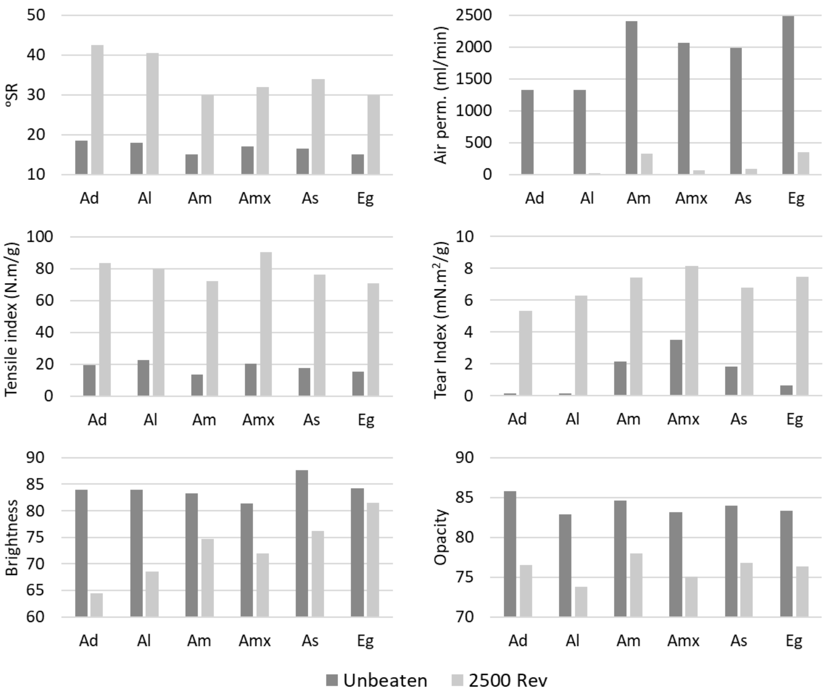

Figure 1. Physical (Schopper-Riegler degree- ºSR, and Air permeability), strength (tensile index and tear index) and optical properties (brightness, opacity) of paper from *A. dealbata* (Ad), *A. longifolia* (Al), *A. mearnsii* (Am), *A. melanoxylon* (Amx), *A. saligna* (As), and comparison to *E. globulus* (Eg).

The Schopper-Riegler degree (°SR) evaluates the drainability resistance of pulp suspensions. This translates into the energy required to increase the pulp's superficial fibrillations and improve the strength of paper properties. *Acacia* species results are within the expected interval of the °SR for unbeaten short fiber pulp. Upon beating, all pulps obtained °SR above 30. *A. dealbata* and *A, longifolia* obtained the °SR above 40, which can be correlated with the fine element content. *A. mearnsii* has a beating behavior similar to *E. globulus*.

The tensile index gives information on fiber strength, bonding and length. Our results showed not much variation between species in unbeaten and beaten pulps. All *Acacia* species presented higher tensile strength than *E. globulus*. Tear index is the strength required to withstand a tearing force and beating increases the tear index for all species.

Regarding optical characteristics, the brightness dropped with beating, being *A. dealbata*, the species in which this fall was higher. This fall was not so pronounced in *E. globulus*. Opacity showed the same behavior. However, the variation between species in the beating pulps was not as high as the brightness.

CONCLUSION

The optimized pulping process (to achieve Kappa number 17) produced pulps with slightly higher yields than those obtained for *E. globulus. A. mearnsii* and *A. melanoxylon* have higher handsheets strength characteristics, making them interesting materials for pulping production. The biggest drawback was that all *Acacia* species lost substantial optical properties upon refining.

Nevertheless, most species studied here seem adequate for pulping and paper end-use, adding to the always-in-need pulpwood supply chain alone or as an addition to the *E. globulus* pulp production. The valorization of wood wastes from invasive control actions helps to promote them and increase their effectiveness in achieving a more sustainable and balanced ecosystem.

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REFERÊNCIAS

1- Richardson DM, Rejmánek M. Trees and shrubs as invasive alien species - a global review. Divers Distrib. 2011;17(5):788–809.

2- Morris TL, Esler KJ, Barger NN, Jacobs SM, Cramer MD. Ecophysiological traits associated with the competitive ability of invasive Australian acacias. Divers Distrib. 2011;17(5):898–910.

3- Le Maitre DC, Gaertner M, Marchante E, Ens EJ, Holmes PM, Pauchard A, et al. Impacts of invasive Australian acacias: Implications for management and restoration. Divers Distrib. 2011;17(5):1015–29.

4- Correia R, Quintela JC, Duarte MP, Gonçalves M. Insights for the valorization of biomass from Portuguese invasive *acacia* spp. in a biorefinery perspective. Forests. 2020;11(12):1–42.

5- Chan JM, Day P, Feely J, Thompson R, Little KM, Norris CH. *Acacia mearnsii* industry overview: current status, key research and development issues. South For. 2015;77(1):19–30.

6- Nambiar, EKS, Harwood, CE, Mendham, DS. Paths to sustainable wood supply to the pulp and paper industry in Indonesia after diseases have forced a change of species from acacia to eucalypts. Australian Forestry. 2018, 81(3):148-161.