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The use of Tagasaste (*Chamaecytisus proliferus*) from different origins for biomass and paper production

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Abstract

In order to identify faster-growing non-woody species usable for biomass and paper production, four Tagasastes (*Chamaecytisus proliferus*) from different origins are tested. All the Tagasaste species (T. Huelva, T. Australia, T. New Zealand and T. La Palma island) show a good soil and climatic adaptation to Southwest Spain. The studied Tagasaste provenances shows biomass productivity ranges from 1.0 t ha^{-1} yr^{-1} to 3.4 t ha^{-1} yr^{-1} (o.d.b.) and 25.3 t ha^{-1} yr^{-1} to 49.4 t ha^{-1} yr^{-1} under Mediterranean conditions for first and second year sprouts, respectively. The quantity of solubles and extractives shows similar values when compared with wood materials. A relatively lower lignin content in Tagasaste (from 13.7% to 17.1%) species has been found with respect to other vegetal species. The α-celullose contents (43.6–45.3%) were in the range of the normal values expected for the other non-wood raw materials.

The study confirms the feasibility of the organocell yield pulping process to Tagasaste provenances. Organocell processes provide an efficient delignification (kappa index from 7.2 to 10.9 and pulp yield from 43.6% to 54.1%). The best results are obtained for the physical properties of paper sheets for Tagasaste from Australia in the second year, with values of tensile index of 16.0 kNm/kg, burst index of 1.12 MPa m^2/kg and tear index of 0.55 Nm^2/kg.

Keywords: Tagasaste; *Chamaecytisus proliferus*; Pulp; Paper; Biomass

1. Introduction

The total global paper-making consumption was projected to increase from 316 million tons in 1999 and 351 million tons in 2005 to about 425 million tons by 2010. The wood consumption was 173 million tons in 2005 (Rowell and Cook, 1998; FAO, 2006). This is in spite of the generalization of computer and audio-visual technologies that could have gradually reduced its consumption. However, an increase in demand for certain types of paper as a result of these technological innovations and environmental demands has occurred (Bayer et al., 1999).

Historically, pulp and paper production has long been recognized as a significant point source of pollution. In recent years, both market demand and environmental pressure have been faced by the pulp and paper industry and to some extent wood has been replaced with non-woody and/or annual plants, and also with agricultural residues (Jiménez et al., 1997). All fibre resources will be required to meet projected demands, including fast growth plantations, increased recycling and non-woody plant fibres from crop residues as well as fibre crops (Alli and Saikia, 1997; Máximo et al., 1998; Cappelletto et al., 1999; Antunes et al., 2000; Gominho et al., 2001).

On the other hand, soil degradation has been described as an important problem in Europe: 12% of total European land area has been affected by water erosion and 4% by wind erosion (Ananda and Herat, 2003). Some of the main effects of soil degradation lead to reduced vegetative cover, decreased water quality, lower efficiency in the use and management of water and an increased risk from pest attack and diseases because of lowered biological control (Zalidis et al., 2000; Paz et al., 2006). The possible solutions

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involve an agricultural sustainability, that using natural resources to progressively enhance the productive capacity without jeopardizing future potential. For example, leguminous species or the ones vaccinated against bacteria have been proposed from the Bradyrhizobium, Rhizobium, Sinorhizobium and Mesorhizobium (Ulrich and Zaspel, 2000; Lafay and Burdon, 2001; Rodríguez-Echevarría et al., 2003), helping the recovery of already degraded grounds. Woody legumes can prevent erosion, increase soil fertility and facilitate the establishment and growth of other plant species (Cross and Schlesinger, 1999; Rodríguez-Echevarría et al., 2003).

Moreover, these crops favourably affect another problem such as the surplus production of food generated by the European Union for the last decade. Three different solutions have been proposed for this situation of surplus production (Montero de Espinosa, 1993): working on very high quality products, though selection of fast-growing species and high profitability (Fernández, 1996), the use of crops that help to recover the degraded environmental balance and leading agriculture to non-food uses (energy, paper, tissues, chemical products, etc.).

The use of Tagasaste or other non-woody faster-growing species for papermaking could have a great advantage in that they provide remediation for the environmental problems associated with deforestation. Tagasaste (*Chamaecytisus proliferus*) began to be researched at on the end of the nineteenth century, but it was only in 1982 that its agronomy features were determined (Snook, 1982a,b; Webb and Shand, 1985; Twosend and Radclife, 1987). Tagasaste grows most easily on sandy soils. It tolerates a low annual rainfall (the lower limit is 300 mm) in some areas of Australia and New Zealand, where this shrub was introduced at the end of the nineteenth century (Méndez and Almeida, 1988).

The objectives of this study were to quantify the effects of four Tagasastes (*Chamaecitysus proliferus*) from different origins: (a) on the yield obtained (total biomass and trimming residues of wood) throughout two years, (b) on the chemical composition changes of the raw materials during that period and (c) on the physico-chemical characteristics of the organocell pulp and paper obtained.

2. Experimental

2.1. Raw material preparation

Plants were obtained from seeds, for four Tagasastes shrubs (*C. Proliferus*) originating from *Huelva*, *Australia*, *New Zealand* and *La Palma island*, which were used in this experiment. These plants were grown in a nursery, in 300 cm^3 pot holders; they were inured to bacterium *Rhizobium* and, when they were three years old, they were planted in the ground in La Rábida (Huelva, south-western Spain).

Field experiments were carried out in two plots with a complete randomized block design with four replicates per provenance. No fertilizers were not added to these plots. The soil at the experimental site was sandy loamy with a pH of 6–8 and had a moderate to substantial depth.

Four samples, representing four Tagasaste provenances aged from 1 to 2 years, and the sprouts of the plant cut again after the first year, were collected (pruning was always done during winter).

Representative foliage and branch wood samples were collected (variety-wise, in quadruplicate) for moisture estimation and chemical analyses, in a random fashion. For yield estimation, four randomly selected plants per plot were cut at the base of the crown. The samples were immediately transferred to the laboratory in double-sealed polyethylene bags. After recording the fresh weights, they were dried to a constant weight at 70 °C, and ground to pass through a 2 mm sieve. Estimates of dry weight biomass were obtained from the fresh weights of various plant types and their corresponding moisture contents. The average biomass of component parts per plant was multiplied by the number of plants per plot and extrapolated to a hectare.

2.2. Characterization of raw material, pulp and paper sheets

Tagasaste wood trimming samples were milled to pass through an 8 mm screen, since no diffusion limitations were observed for the particle size in preliminary studies. Samples were air-dried, homogenized in a single lot to avoid differences in composition among aliquots, and stored.

Aliquots from the homogenized wood lot were subjected to moisture determination (drying at 105 °C to a constant weight) and to quantitative acid hydrolysis with 72% sulphuric acid following standard methods (Browning, 1967). The solid residue after hydrolysis was recovered by filtration and considered as Klason lignin.

The characterization experiment involved the following parameters: 1% NaOH solubles (Tappi 212), hot water solubles (Tappi 204), ethanol–benzene extractives (Tappi 204), α-cellulose (Tappi 203-OS-61), and holocellulose (Wise et al., 1946). All treatments in this study were in a completely randomized design with five replications (coefficient of variation less than 5%).

For the determination of fibre length, 100 individual fibres were measured from each variety. Statistical analyses were performed using ANOVA and the differences among varieties were compared using Tukey's test. The means were separated on the basis of the least significant difference at the 0.05 probability level.

Tagasaste wood trimmings were used for pulp and papermaking, but only wood was considered as it contained the bark, which was very thin and difficult to strip off. This accounted for only 1–2% of the overall mass.

Characterization experiments of pulp involved the following parameters: yield (Tappi 257), kappa index (Tappi 236) and viscosity (UNE 57-039). From paper sheets, we determined grammage (UNE 57-014-74), tensile index

(UNE 57054 and UNE 57-028), burst index (UNE 57058) and tear index (UNE 57-033-86).

2.3. Pulping and papermaking

Celullose pulps were obtained using a 4-L cylindrical reactor bath that was heated by means of electrical resistances and linked to a control unit including the required instrument for measurement and control of pressure and temperature. The control unit included temperature and pressure gauges as well as appropriate safety devices. The initial liquor to solid ratio was 8:1 (dry wt. basis); the aqueous soda concentration in the cooking liquor was 21% by weight; the ethanol concentration was 30% in volume and the anthraquinone concentration was 0.05% in weight. The reactor was then closed and simultaneously heated and activated to assure good mixing and uniform swelling of the wood. The temperature was set at 185 °C for 60 min and preheating was done for 30 min to reach the temperature mentioned. Finally, to open the reactor, the liquor was quickly refrigerated by an internal heat exchanger to obtain low-pressure levels. Following cooking, the pulp was separated from the liquor and disintegrated, without disturbing the fibres for 3 min (2500 rpm), washed on a sieve of a 16 mm mesh, defibered on a Sprout-Waldron refiner and passed again thought a Strainer filter (0.4 mm mesh) to isolate the uncooked material. The selection of the process and variables was based on bibliographic references (from different authors and raw materials) and previous experiences (Díaz et al., 2004; López et al., 2004).

Paper sheets were prepared with an ENJO-F-39.71 sheet machine according to the UNE 57042-74 standard.

3. Results and discussion

3.1. Biomass production

All Tagasastes from different origins show a good soil and climatic adaptation to the zone. Biomass accumulation shows important variations (Table 1).

With respect to the kind of exploitation (no of pruning), the reappearance after the pruning in the first year was not strong. We used the expression "$1 + 1$" for denoting this reappearance. Some plants (30%) withered and, as a whole, the production was decreased significantly. The reason could be that the height of the pruning was not enough or that these plants should have been more developed. Within the second year, the growth of the plants which were not pruned was very high, and was comparable to the growth experimented by some species of plants such as Leucaena, which grow up quickly (Díaz et al., 2007).

When pruned for two years, those plants picked up in their natural habitat (La Palma Island) were the least productive, differing from those plants that came from improved origins (New Zealand, Huelva and Australia). Plants coming from such origins were also not productive in the reappearing $(1 + 1)$. Notwithstanding this, their growth was more than the average growth during the first year but it was not significant; taking into account absolute figures, the one year production is very sparse compared with the two year production (Table 1).

With regard to the distribution of biomass, neither the effects of the plot nor the effects of the block were significant, so it was omitted from the analysis. The one year $(1 + 1)$ reappearance of the four origins has a diameter, height and biomass inferior to that of the two year plant, which showed a considerably larger non woody biomass ratio. The pruning stunted the growth of the plant and decreased its biomass to between 10% and 50% compared with the two year plant. The two year plants from New Zealand had the largest diameter, height and their biomass was more than three times the biomass of the other plants.

The production of Tagasaste dry material obtained is similar to the highest production obtained from other alternative crops such as *kenaf, miscanthus or sorghum* (Montero de Espinosa, 1993; Junta de Extremadura, 1996) in Spain and exceeds those obtained for tree species such as poplar, robinia, willow, eucalyptus or ailanthus, which never exceed 20 t ha^{-1} yr^{-1} (poplar trees) (Hernández et al., 1996). They fix nitrogen from the atmosphere and

Table 1
Biomass yielded and characteristics of fibre from Tagasaste from different origins for two years

	Pruning to year 1		Pruning to year 2		Year 1 sprout	
	WD[A]	TD[B]	WD[A]	TD[B]	WD[A]	TD[B]
Huelva	1.45(0.47)	2.12(0.58)	39.45(8.43)	49.36(9.14)	1.51(0.57)	1.90(0.74)
Australia	0.57(0.26)	1.04(0.44)	32.49(4.51)	42.23(8.03)	0.58(0.37)	0.81(0.54)
New Zealand	2.16(0.78)	3.37(1.24)	38.75(3.41)	49.04(3.50)	2.36(0.60)	3.00(0.93)
La Palma Island	1.32(0.36)	2.29(0.60)	17.80(5.30)	25.32(6.80)	0.23(0.12)	0.32(0.17)
	Height	Diameter	Height	Diameter	Height	Diameter
Huelva	1.19a	1.83a	2.99c	2.08b	2.30b	1.80a
Australia	1.05a	1.00a	2.94b	2.31b	1.46a	1.23a
New Zealand	1.13a	1.44a	3.52b	3.50b	2.06a	1.80a
La Palma Island	1.23a	1.89a	2.58b	2.11b	1.58a	1.94a

Values followed by the same letter in the same column do not differ significantly ($p < 0.05$).
[A] WD: dry wood (t ha^{-1} yr^{-1}).
[B] TD: total dry biomass (t ha^{-1} yr^{-1}).

they have multiple uses (e.g. forage production). They are an interesting alternative for crops for soil recovery with industrial and livestock use.

3.2. Characterization of raw materials

The physico-chemical characterics for Tagasaste varieties for the first year (and year 1 sprout) and the second year are shown in Table 2. The fibre length of all studied Tagasastes is not statistically different between the first and the second year harvest, except for the one from *Australia* which suffers an increase of 12.3%. The Tagasaste from Huelva has the smallest fibre length (0.55 mm), in comparison with the other studied Tagasastes, whose fibre lengths are between 0.74 and 0.83 mm.

If they are analyzed as a whole, the characteristics related to the predictable yield in cellulose pulp (hot water, 1% NaOH soluble and ethanol–benzene extractives) imply a higher potential for second year pruning if they reduce hot water solubles between 43% and 58.5%, and ethanol–benzene extractives between 2% and 24%.

The holocellulose content of the Tagasates from different origins under investigation was higher than 75%, with respect to o.d. material, meaning that the cellulose/hemicelullose ratio was higher than 1.2, which is common to other vegetal species. This ratio is very important if one considers the capital role that hemicelullose plays in papermaking (Cordeiro et al., 2004). Holocellulose content becomes higher from the first to the second year harvest, except for the Tagasaste from Australia which decreases. Nevertheless, the α-cellulose/holocellulose ratio decreases in all the species studied, but not in the Huelva one, which remains constant.

The lignin content for Tagasastes from different origins was lower than 17.1%. Tagasastes from Huelva and New Zealand show a lower lignin content for the first year. However, the opposite occurs for the Tagasaste from Australia and La Palma.

Table 3 shows the chemical characterization of the other bibliographic raw materials (hardwoods, softwoods and alternative raw materials). The quantity of hot water and 1% NaOH and ethanol–benzene extractives shows similar values when compared with wood materials and their values are relatively lower when they are compared with non-woods and some other annual plants. Pulp yields are negatively correlated with the extractive content (ethanol-benzene, water soluble); therefore a greater paste yield could be supposed for these Tagasastes from different origins.

A relatively lower lignin content in Tagasaste from different origins has been found with respect to other vegetal species (Table 4 and Ververis et al., 2004). The low values lignin content observed in Tagasastes from different origins, could suggest that they may require a low pulping time and chemical charge compared with hardwoods, softwoods and those of the other non-wood raw materials.

On the other hand, the holocellulose content shows similar values when compared with hardwoods and their values are relatively higher when they are compared with softwoods and most of non-woods (Table 3).

The α-cellulose content found in Tagasates from different origins are in the range of the normal values expected for other non-wood raw materials and lower than those found for wood-based materials.

3.3. Characterization of pulps and papers

Industrial use has been considered by means of celullose pulp and paper production through tests described in the Section 2.

In Table 4, the values found for the physico-chemical characterization of the pulps and paper sheets obtained for the different origins of Tagasastes, in the first year (and sprouts, after prunings, with in one year) and the second year, are shown.

These results are comparable to other alternative crops and better than the other tree species such as eucalyptus. Much information exists about the state of the art. For example, with respect to eucalyptus, Botello et al. (1999) reports a yield of 50.5% using the ethanol process. In sev-

Table 2
Physico-chemical characterization of the first year (and sprouts, after pruning, within one year) and the second year Tagasaste from different origins

	Fibre length (mm)	1% NaOH solubles (%)	Ethanol–benzene extractives (%)	Hot water solubles (%)	Holocellulose (%)	Lignin (%)	α-Cellulose (%)
First year and sprouts within one year							
T. Huelva	0.55	16.7	2.6	2.8	79.7	16.8	45.4
T. Australia	0.74	15.6	2.2	3.0	82.2	15.7	47.7
T. New Zealand	0.79	16.2	3.4	3.0	75.4	14.8	43.6
T. La Palma	0.75	16.6	3.3	2.4	76.5	14.1	45.0
Second year							
T. Huelva	0.55	14.0	2.6	1.6	80.1	16.0	45.8
T. Australia	0.83	14.3	1.8	1.7	80.8	16.6	45.1
T. New Zealand	0.79	17.0	2.6	1.2	80.0	13.7	43.9
T. La Palma	0.72	16.7	2.8	1.1	82.2	17.1	46.0

Percentages with respect to initial raw material (100 kg o.d.b.).
Values followed by the same letter in the same column do not differ significantly ($p < 0.05$).

Table 3
Physico-chemical characterization of some raw material References from others authors

Raw material	Holocellulose (%)	Lignin (%)	α-cellulose (%)	Hot water solubles (%)	Ethanol–benzene extractibles (%)	1% NaOH solubles (%)	References
Word materials							
Eucalyptus globulus	80.5	20.0	52.8	2.8	1.2	12.4	Alonso (1976)
Eucalyptus globulus	79.5	21.2		2.9	1.4	12.8	Gilarranz et al. (1999)
Eucalyptus globulus	72.6	22.9	46.6				Parajó et al. (2004), Garrote and Parajó (2002)
Beech (*Fagus sylvatica*)	62.11	23.7	38.6		0.49		Dapía et al. (2002)
Pinus pinaster	60.5	30.2	42.9				Parajó et al. (1993)
Pinus pinea	69.6	26.2	55.9	2.0	2.6	8.0	Alonso (1976)
Aspen wood	68.3	21.4	47	0.39	0.64		Abad et al. (2000), Yañez et al. (2000)
Non-wood materials							
Cannabis sativa L. (hemp).		21.8	37.3				Antunes et al. (2000)
Cynara cardunculus L.	63.4	19.6	38.0	13.0	5.0		Antunes et al. (2000)
Gossypium hirsutum L. (cotton)	72.9	21.5	42.3	3.3	1.4	20.3	Jiménez et al. (1993)
Hibiscus cannabinus L.(kenaf)				6.7	2.0	26.1	Khristova et al. (2002)
Panicum virgatum L. (switchgrass)	78.5	18.1		15.1	3.7	45.4	Law et al. (2001)
Panicum virgatum L. (switchgrass)	81.0	19.5		12.4	1.7	27.9	Law et al. (2001)
Triticum sp. (wheat straw)	70.7	21.7	41.3		2.5		Sun and Tomkinson (2004)
Olea europaea (olive)	69.1	17.6	41.0	17.3	12.2	30.0	Díaz et al. (2005)

Table 4
Physico-chemical characterization of the first year (and sprouts, after pruning, within one year) and the second year Tagasastes from different origins pulp obtained

	Yield (%)	Kappa index	Viscosity (cm^3/g)	Tensile index (kNm/kg)	Burst index (MPa m^2/kg)	Tear index (Nm^2/kg)
First year and sprouts within one year						
T. Huelva	43.6	10.9	544	13.7	0.53	0.96
T. Australia	44.3	8.3	631	14.8	0.50	0.78
T. New Zealand	47.1	8.3	672	14.3	0.55	0.98
T. La Palma	47.6	9.1	640	13.0	0.44	0.34
Second year						
T. Huelva	46.3	7.5	513	12.1	0.45	0.56
T. Australia	48.1	8.9	736	16.0	0.55	1.12
T. New Zealand	50.3	7.2	629	13.6	0.52	0.89
T. La Palma	52.1	8.1	755	13.4	0.45	0.95

eral articles there appear kappa numbers between 20 and 110 (Lora, 1992; Pascoal Neto and Robert, 1992). Pereira et al., 1986 reports rupture length values between 1.4 and 1.7 km for ethanol pulping of *Eucalyptus globulus* and 1.3 for the kraft pulping process without refining (tensile index: 13–17 kNm/kg approximately).

Cellulosic pulp yield from Tagasaste improves in the second year, but is not enough (specially, when compared to others species) and it never reaches more than 10% with respect to the pulp yield in the first year. The higher yields that have been found are 52.1% for Tagasaste from La Palma and 50.3% for Tagasaste from New Zealand.

It can be seen in Table 4 that values for the kappa index are very low for all Tagasaste species, from both harvests (first and second years). They never reach a value above 11, which suggests they have a good tendancy to whiten. The lowest values for the kappa index are for New Zealand and Huelva in the second year (7.2 and 7.5, respectively).

Tagasastes from Huelva and New Zealand exhibit better tensile index, burst index and tear index for paper sheets in the first year. However, the opposite occurs for the Tagasaste from Australia and La Palma. Better results are obtained for physical properties of paper sheets for Tagasate from Australia in the second year, with a tensile index of 16.0 kNm/kg, burst index of 1.12 MPa m^2/kg and tear index of 0.55 Nm^2/kg. Regarding sheet viscosity, similar effects are found with higher viscosity values for Tagasaste from Australia (736 cm^3/g) and La Palma (755 cm^3/g) for the second year.

4. Conclusions

All the Tagasastes from different origins (T. Huelva, T. Australia, T. New Zealand and T. La Palma island) showed a good soil and climatic adaptation to the Southwest of Spain. Due to their nitrogen fixing property and their multiple uses they are an interesting alternative for crops for ground recovery and for their agricultural and industrial use. In respect to the kind of exploitation (no of pruning), within the second year, the growth of the plants which were not pruned was very high (from 25.3 t ha^{-1} yr^{-1} – T. La Palma island – to 49.4 t ha^{-1} yr^{-1} – T. Huelva). These productions are comparable to other alternative crops such as *Kenaf*, *Miscanthus* or *Sorghum* in Spain and better than other tree species such as poplar, robina, willow tree, eucalyptus or ailanthus.

No statistically significant differences in fibre lengths have been found for Tagasastes from different origins (0.724–0.833 mm) except for the lower values found for Tagasaste from Huelva (0.55 mm).

The quantity of hot water and 1% NaOH solubles and ethanol–benzene extractives shows similar values when compared with wood materials and their values are relatively lower when compared with non-wood and some other annual plants. A relatively lower lignin content in Tagasaste (13.7–17.1%) species has been found with respect to other vegetal species. The α-cellulose contents found in origin Tagasaste are in the range of the normal values expected for the other non-wood raw materials and lower than those found for wood-based materials.

Cellulosic pulp yield from Tagasaste improves the second year, but is not enough (specially, when compared to others species) and it never reaches more than 10% with respect to the pulp yield in the first year. On the whole, the values for the kappa index are quite low for all Tagasastes, for both first and second year harvests, as they never reach a value above 11, which suggests their good tendency to whiten.

In the first year, Tagasastes from Huelva and New Zealand exhibit better values for tensile index, burst index and tear index for paper sheets. However, the opposite occurs for the Tagasaste from Australia and La Palma island.

The Tagasaste from Australia is the best, based on because the physical characteristics of its paper sheets (tensile index: 16.0 kNm/kg, burst index: 0.55 MPa m^2/kg and tear index 1.12 Nm^2/kg) for the second year harvest.

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