PREPARATION OF PAPER SHEETS FROM CELLULOSIC FIBRES OBTAINED FROM *PRUNUS AMYGDALUS* AND *TAMARISK* SP.

NABAWIA MECHI, RAMZI KHIARI,[*,**,***] ELIMAME ELALOUI and MOHAMED NACEUR BELGACEM[**,***]

Materials, Environment and Energy Laboratory, Faculty of Sciences, University of Gafsa, Tunisia
[*]*University of Monastir, Faculty of Sciences, UR13ES63-Research Unity of Applied Chemistry & Environment, 5000 Monastir, Tunisia*
[**]*University of Grenoble Alpes, LGP2, F-38000 Grenoble, France*
[***]*CNRS, LGP2, F-38000 Grenoble, France*
✉ *Corresponding author: Ramzi Khiari, khiari_ramzi2000@yahoo.fr*

Received April 17, 2015

This work is devoted to the valorization of two residues: *Prunus amygdalus* and *Tamarisk* sp. First, their chemical compositions were studied according to standard methods. The results revealed that the existing amounts of holocellulose, lignin and cellulose were close to those encountered in other annual plants and non-wood species. The amount of extractives in different solvents and the ash contents were found to be relatively high. The second part of this work focuses on the use of a classical soda-anthraquinone process to produce cellulosic fibres from two plants. The ensuing fibres were characterized by several methods, such as FT-IR analysis, ATG-DTG and X-ray diffraction (XRD). Moreover, the cellulosic fibres arising from these two plants were used successfully in the preparation of paper sheets. The mechanical properties, air permeability and morphology of the prepared materials were assessed. Furthermore, the pulps exhibited good drainage ability, while the ensuing papers excellent mechanical properties. Thus, these agricultural by-products were found suitable for valorization into cellulose fibres, for papermaking applications, and could also be used to obtain cellulose derivatives and/or fibre-reinforced composite materials.

Keywords: *Tamarisk* sp., *Prunus amygdalus*, chemical composition, pulping, papermaking

INTRODUCTION

Over the last decades, the use of cellulosic fibres has intensified, which increased the market demand of such raw material. Thus, to satisfy the large demand of natural fibres, it becomes challenging to supply the needs of all the users at reasonable costs, with required quantities and qualities. In fact, cellulosic fibres are already widely used for many applications. They are used in textile industries (to manufacture clothing and furniture), papermaking and packaging industries,[1-3] in pharmaceutical areas (compresses, dressings, bandages, excipient, drugs etc.) and for the preparation of innovative materials such as bio-composites. Consequently, the consumption of cellulosic fibres is increasing, and it is becoming increasingly difficult to satisfy the large request. In this context, non-wood species or annual plants can be viewed as alternative sources of cellulosic fibres, especially in regions that are poor in forest resources. Non-wood fibres could be potentially used in applications that require materials with similar properties as those provided by wood fibres. Finally, non-wood fibres are often obtained from agricultural waste and can therefore be valorised, which fits very well with the actual context, so-called "circular economy".[1,5-7] From this perspective, it is important to valorise two lignocellulosic materials largely available in Tunisia, *Prunus amygdalus* and *Tamarisk* sp., as a source of cellulosic fibres (Fig. 1). In fact, these are two of the most cultivated plants in arid and semi-arid regions of the world,[8-12] in general, and in Tunisia, in particular.

Significant quantities of *Tamarisk* sp. trunk waste are generated every year on Tunisian agricultural lands. *Prunus amygdalus* is also an agricultural residue, known as almond plant, and produced mainly in the USA, Spain, Iran, Syria, Italy, Morocco, Turkey and Tunisia with an

amount of about 2 million tons in 2012 (FAOSTAT data). According to FAO (FAOSTAT data), Tunisia produces *Prunus amygdalus* with 3.8% capacity.[13] In Tunisia, this plant is cultivated on about 190000 hectares (FAOSTAT data). To the best of our knowledge, no data about the chemical composition of *Prunus amygdalus* and *Tamarisk* sp. are available in the literature. The almond and *Tamarisk* sp. cultures produce a huge amount of wastes, which are left to biodegrade and fertilise agricultural lands. The present work deals with the study of these two abundant renewable resources, namely: (i) their chemical composition; (ii) their soda-anthraquinone cooking; (iii) the characterisation of the obtained pulps and (iv) their valorisation in papermaking applications. Thus, the morphology of the isolated fibres was studied. Then, these fibres were used to prepare paper sheets and the ensuing materials were characterised in terms of physical and mechanical properties of the prepared handsheets. The obtained results were discussed and compared with those available in the literature related to wood-, non-wood- and annual plant crops-based papers.

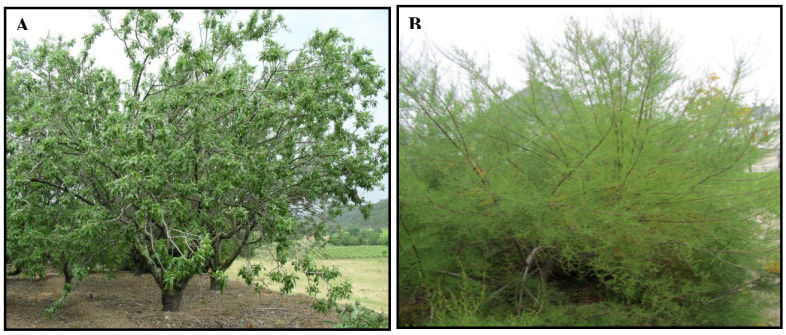


Figure 1: *Prunus amygdalus* (A) and *Tamarisk* sp. (B) trees

EXPERIMENTAL

Raw material

Prunus amygdalus and *Tamarisk* sp. were obtained from Gafsa-Tunisia in September 2014 and dried under natural conditions (relative humidity (RH) of 65% and average temperature around 25 °C). They were then washed in order to eliminate sand and other contaminations and dried again under the same conditions. Before pulping, the investigated raw materials (*Prunus amygdalus* and *Tamarisk* sp.) were cut into small pieces with lengths of about 1–3 cm in order to facilitate the extraction of fibres. All the chemicals were purchased from Sigma-Aldrich and were used without further purification.

Chemical composition

The chemical composition of *Prunus amygdalus* and *Tamarisk* sp. was determined in terms of the amount of extractive substances in 1% sodium hydroxide and ethanol-toluene mixture (20/80 v/v). Ashes, Klason lignin, holocellulose and α-cellulose were determined using the standard methods listed in Table 1. All the measurements were carried out at least in triplicate and the experimental error was within 5%.

Pulping and characterization of suspension fibres

Cooking process

The pulping of *Prunus amygdalus* and *Tamarisk* sp. was carried out according to a method derived from that described by Khiari *et al.*[14] Briefly, sixteen grams of *Tamarisk* sp. or *Prunus amygdalus* were cooked at 160 °C, with a total alkali charge of 20% expressed in NaOH (w/w, based on oven dried (o.d.) material), an anthraquinone concentration of 0.1% (w/w, with respect to o.d. material) and a cooking time of 120 min. The resulting fibres were then washed extensively with water until a neutral pH was acquired. Finally, the fibres were dried (under atmospheric conditions: 25 °C and 50% RH). All the cooking experiments were repeated at least in triplicate.

Characterization of extracted fibres

Elemental composition

The analysis of elemental composition of the obtained fibres from *Prunus amygdalus* and *Tamarisk* sp. was investigated using scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS). The EDS spectra revealed the different atoms present in the sample under investigation.

Pulp characterization

After cooking, the extracted fibres were separated from the black liquor, washed with water until a clear filtrate was obtained. The fibre suspension was then disintegrated according to the standard method ISO 5263-1. This operation allowed the removal of the uncooked materials by passing the diluted pulp suspension through a slotted screen of 0.15 mm aperture size. The resulting pulps were characterised in terms of yield and degree of polymerization. The

cooking yield was determined as the ratio of weight of oven dried (o.d.) material after washing to that of the initial raw material. The degree of polymerization (DP) was evaluated using the Tappi methods (T 230 om-99) for which the following equation (1) was used:[15]

$$DP_v = [0.75\ (954 Log_{10}\eta\text{-}325)]^{1.105} \quad (1)$$

In all cases, the quantity of residual lignin was too low, so that no corrections were applied to Eq. 1.

The morphological properties of the prepared fibres were studied using a MORFI analyser. The main parameters of the fibre (length, width, % of fine elements) were evaluated by image analysis of a diluted suspension flowing in a transparent flat channel observed by a CCD video-camera. The test was repeated at least in triplicate.

The water retention value (WRV) of the pulp was determined according to the method reported by Silvy *et al.*[16] Briefly, this method consisted in centrifuging wet pulp samples for 15 min at 3000 g. Before and after drying, the samples were weighed and WRV was calculated by using the following equation:

$$WRV(\%) = 100 \times \frac{(M_1 - M_2)}{M_2} \quad (2)$$

where M_1 is the mass of the wet sample after centrifugation and M_2 is that after drying of the wet sample at 105 °C to a constant weight. The pulp drainage ability was evaluated by measuring the Schopper-Riegler degree (SR–ISO 5267-1).

Amount of carboxylic groups

The total amount of carboxylic groups in both pulps from *Prunus amygdalus* and *Tamarisk* sp. was assessed by conductometric titrations according to the procedure described by Katz *et al.*[17] Before the titration, the investigated fibres (0.6 to 1 g in 300 mL of deionized water) were acidified by adding 1 mL of HCl solution (0.1 mol.L^{-1}) and extensively washed. The conductivity of the fibre suspension was then adjusted to 600 µS.cm^{-1} with a 0.5 mol.L^{-1} NaCl solution. Finally, the titration of 500 mL of fibre suspensions was performed with a NaOH solution (0.01 mol.L^{-1}), after the addition of 0.5 mL of a solution of HCl (0.1 mol.L^{-1}).

Thermal stability of the prepared fibres

The thermal stability of the extracted fibres was characterized using a TA instrument Q-50 thermogravimetric analyzer (TA instruments, USA). The samples were heated under nitrogen flow from 30 to 800 °C, at a heating rate of 10 °C.min^{-1}.

Infrared spectroscopy (FT-IR-ATR)

The FT-IR analysis of the prepared fibres was carried out with a Perkin-Elmer 1000 spectrometer equipped with a diamond ATR accessory. The spectra were obtained in the wavenumber range of 600-4000 cm^{-1} with 32 scans.

X-ray diffraction pattern (XRD)

The crystallinity of the extracted materials was studied by X-ray diffraction (XRD). Each sample in the form of milled powder was placed on the sample holder and levelled off to obtain total and uniform X-ray exposure. The ensuing samples were then examined using an X-ray diffractometer (D8-Advance Bruker AXS GmbH) at room temperature with a monochromatic CuKα radiation source (λ = 0.154 nm) in the step-scan mode with a 2θ angle ranging from 5° to 60° with a step of 0.04 and a scanning time of 5.0 min. During this work, the method described by Segal *et al.*[18] was used in order to evaluate the crystallinity of the extracted fibres. The crystallinity index CI was determined based on the reflected intensity data following Eq. 3:

$$C_I(\%) = 100 * (\frac{I_{002} - I_{am}}{I_{002}}) \quad (3)$$

where I_{am} is the intensity scattered by the amorphous part of the sample and I_{002} – the maximum intensity of the (002) lattice diffraction peak. The diffraction peak for plane (002) is located at a diffraction angle around 2θ = 22° and the intensity scattered by the amorphous part is measured as the lowest intensity at a diffraction angle around 2θ = 18°.

Preparation of paper sheets

First, the fibre suspensions were diluted to 2 g.L^{-1}. Then, conventional handsheets with a basis weight of 60 g.m^{-2} were prepared using a Rapid Kothen sheet former and according to the standard method ISO 5269-2 and adapted for unrefined virgin fibres. The ensuing handsheets were conditioned (23 °C, 50% relative humidity –ISO 187) before testing. The structural and mechanical properties were determined by measuring basis weight, thickness, bulk and permeability, as well as the tensile, burst and tear strength according to their respective standards ISO536, ISO 534, ISO 5636-3, ISO 1924-3, ISO 2758 and ISO 1974.

RESULTS AND DISCUSSION

Chemical composition

The chemical composition of the Tunisian *Prunus amygdalus* and *Tamarisk* sp. was established, as listed in Table 1. The following remarks can be made based on the data from this table:

(i) The main difference between *Prunus amygdalus* and *Tamarisk* sp. is related to cold water (and hot water) extractives content, which is much higher for *Tamarisk* sp., *i.e.* 22.3% (25.4%) compared to that of *Prunus amygdalus*, *i.e.* 11.3% (12.3%);

(ii) The amounts of extractives in ethanol-toluene solvent mixture and the ash contents are

comparable to those reported for other annual plants or agricultural crops[19-23] and wood;[14,24,25]

(iii) Klason lignin was found around 19%, which is in the range of that found for other annual plants;[6,7]

(iv) The holocellulose and α-cellulose contents are similar for both raw materials studied (38.9% for *Tamarisk* sp. and 40.7% for *Prunus amygdalus*). Thus, the polysaccharide content is close to that known for wood materials, which justifies the cooking of *Prunus amygdalus* and *Tamarisk* sp.

Table 1 also summarizes the chemical composition data, as reported in the literature, for two Tunisian cellulosic biomasses, such as date palm rachis and *Posidonia oceanica* balls.[14] The comparison with the present work leads to several comments. Thus, in cold and hot water, the quantity of extractives for both *Prunus amygdalus* and *Tamarisk* sp. is higher than those found in hardwood and softwood, but comparable to those usually contained in non-wood sources.[14,19-22,24,26] The extractives in 1% NaOH (16.8% for *Tamarisk* sp. and 28.7% for *Prunus amygdalus*) are similar to those of wood sources, *i.e.* less than 20%, but are lower than those known for other common annual plants. Finally, the amount of holocellulose for the raw materials under investigation is relatively high, although in the same order of magnitude as those observed for other annual plants or agricultural crops.[19-23] Regarding the ash substances, their content is high for *Tamarisk* sp. (10.7%) and comparable to that of *Posidonia oceanica* balls, rice dishes, Banana pseudo-stems and Amaranth, which exhibited the highest contents, as already reported in the literature.[14] This can be attributed to a pollution of the investigated material by sand, even if intensive washing was performed before proceeding to the characterisation of this raw material. On the opposite, the ash content in *Prunus amygdalus* (3.6%) is lower even if it remains at the same order of magnitude as some of the non-wood plants, such as date palm rachis *etc*.[14]

Table 1
Chemical composition of Tunisian *Prunus amygdalus* and *Tamarisk* sp. – comparison with data collected from previously published studies

Amounts in % (w/w with respect to o.d. raw material)	*Prunus amygdalus*	*Tamarisk* sp.	Date palm rachis[14]	*Posidonia oceanica* balls[14]
Cold water extractives, %	11.3	22.3	5.0	7.3
Hot water extractives, %	12.3	25.4	8.1	12.2
1% NaOH extractives, %	28.7	16.8	20.8	16.5
Ethanol-toluene extractives, %	5.0	3.33	6.3	10.7
Ash, %	3.6	10.7	5	12
Lignin, %	19.2	19.8	27.2	29.8
Holocellulose, %	60.7	58.2	74.8	61.8
Cellulose, %	40.7	38.9	45	40

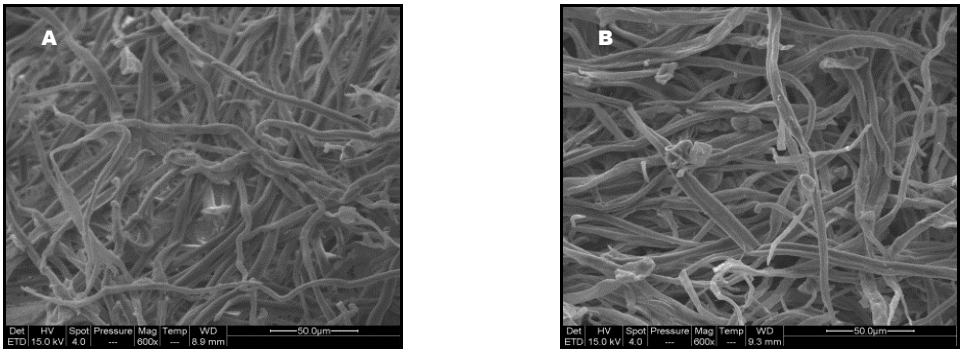


Figure 2: SEM images of *Prunus amygdalus* (A) and *Tamarisk* sp. (B) pulps

Table 2
Main properties of *Prunus amygdalus* and *Tamarisk* sp. pulps

	Prunus amygdalus	*Tamarisk* sp.	Date palm rachis[14]	*P. oceanica* balls[14]
Total akali charge expressed in NaOH, %	20	20	20	20
Anthraquinone concentration	0.1	0.1	0.1	0.1
Time at constant temperature, min	120	120	120	120
Temperature, °C	160	160	160	160
Cooking yield, %	45.2	41.7	44.8	63.6
Screening yield, %	95	94	94	96
Pulp viscosity (mPa.s)	5.28	5.03	15.7	5.4
DP (pulp)	405	386	1203	513
Schopper Riegler (°SR)	17	21	14	10
WRV % (w/w on o.d. pulp)	61.6	61.6	138	110
Fibre length (mm)	0.480	0.369	0.89	0.55
Fibre width (μm)	21	20	22.3	21.3
Fine elements (% in length)	24.3	34.3	30.8	7.5
Total charge	110	100	291	-

Pulping evaluation

Soda-anthraquinone pulping was carried out in a rotating system consisting of 6 reactors with a capacity of 1L each. They were heated electrically, under controlled temperature. The analysis of the ensuing pulp (Fig. 2), in terms of Schopper Riegler, fibre morphology and water retention values, are presented in Table 2. The cooking yields were 41.7 and 45.2% for *Tamarisk* sp. and *Prunus amygdalus*, respectively, which is typically observed for the majority of the non-wood plants.[14,19-24]

Regarding the morphology, it can be noticed that the width of the fibres from *Prunus amygdalus* and *Tamarisk* sp. is close to that of other fibres isolated from several annual plants, while their average length (0.4 mm and 0.3 mm) is significantly lower. These properties impact the aspect ratio, whose value is 20 and 21, for *Prunus amygdalus* and *Tamarisk* sp., respectively. These values are similar to those established for *P. chloranthus* pulp (16).[7] Furthermore, the two investigated fibres have relatively low water retention value (WRV) around 61%, which is lower than that of softwood or hardwood pulps (*ca*. 100%) or some annual plants, such as *Posidonia oceanica* balls (110%)[14] and date palm rachis (138%).[14] *Prunus amygdalus* and *Tamarisk* sp. pulp exhibited lower drainage ability expressed in terms of Schopper Riegler (°SR) degree. In spite of their very high content of fine elements, the SR of these pulps (21 and 17 for *Tamarisk* sp. and *Prunus amygdalus*, respectively) is similar to that of unrefined softwood pulps and lower than that of other non-wood sources and annual plants like *Cynara cardunculus L.* (25)[19,27] and *Miscanthus sinensis* (14).[28]

As presented in Table 2, the total charge borne by the investigated pulps shows that the *Prunus amygdalus* and *Tamarisk* sp. fibres have similar values, *i.e.* 110 and 100 $\mu eq.g^{-1}$, respectively. These values are very similar to those known for other lignocellulosic pulps from other annual plants (typically around 100). This parameter is a function of the cooking yield. Thus, for example, in the case of unbleached kraft pulp from spruce with yields of 49, 48 and 44%, the total charge decreased accordingly, *i.e.*, 139, 91 and 54 $\mu eq.g^{-1}$, respectively.[29] Unfortunately, very few data related to the characterization of non-wood pulps are available in the literature, which makes impossible any reliable comparison.

Finally, the DP of the *Prunus amygdalus* and *Tamarisk* sp. fibres are around 405 and 386 respectively, which are lower than those obtained for unbleached kraft wood fibres (generally about 1300-1500). Nevertheless, these values are comparable to those found for *Posidonia oceanica* (around 513). This result predicts that lower strength properties of the prepared papers should be expected, but opens an avenue for using these substrates in the area of cellulose derivatives.

FT-IR

Figure 3 shows the FTIR spectra of *Prunus amygdalus* and *Tamarisk* sp. fibres. These spectra display several absorption bands, namely: (i) a very broad band at 3340 cm^{-1} representing the O–

H stretching of cellulose and water, a strong band at 2910 cm^{-1} typical of C–H stretching, and others in the region of 1800-600 cm^{-1} accompanying cellulosic structural units. These characteristic bands are analogous to those reported in the literature for other lignocellulosic fibres: 1730 cm^{-1} (C=O ester stretching vibration); 1638 cm^{-1} (C=O aldehyde stretching vibration and also adsorbed/absorbed water molecules); 1420 cm^{-1} (aliphatic C–H vibration); 1030 cm^{-1} (C–O–C stretching vibration of ether groups). The presence of hemicelluloses is hard to distinguish, since the main signals associated with them are overlapped with those of cellulose (polysaccharides). However, the residual lignin content is too low (less than 2%), so that the associated signals are quite weak. Nevertheless, two bands at 1350 and 1220 cm^{-1} are observed, which indicate the presence of a small amount of syringyl groups belonging to lignin macromolecules.

Thermal analysis of pulps from* Prunus amygdalus *and* Tamarisk *sp.

The thermal analysis of the pulps from *Prunus Amygdalus* and *Tamarisk* sp. was performed, and the results displayed in Figure 4 indicate that both pulps behaved similarly. In fact, even if *Tamarisk* sp. had better heat resistance than *Prunus amygdalus*, no significant difference could be pointed out. Moreover, the thermal degradation of these fibres can be divided into three thermal stages: below 250 °C, between 300 to 450 °C and higher than 450 °C.

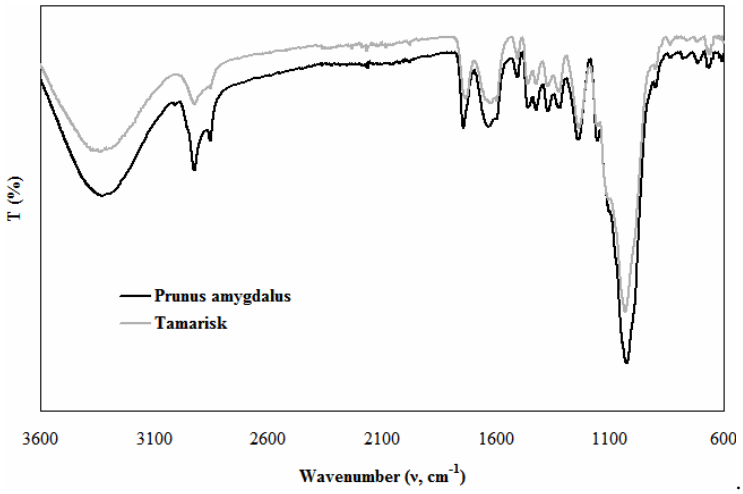


Figure 3: FT-IR spectra corresponding to *Prunus amygdalus* and *Tamarisk* sp. fibres

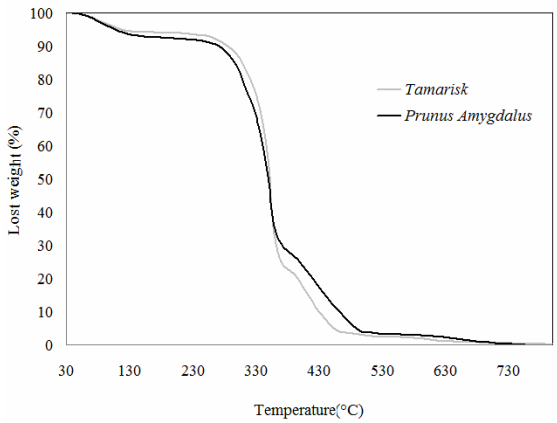
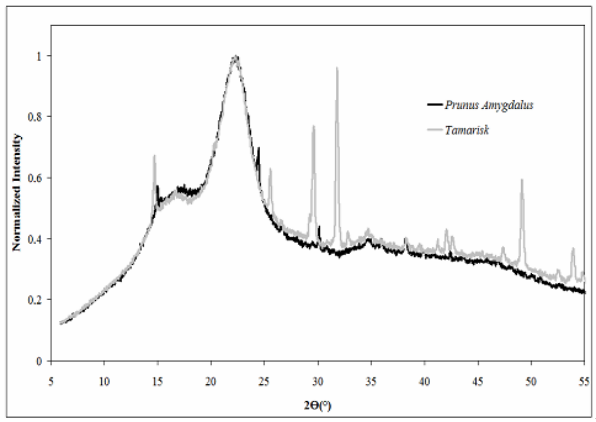
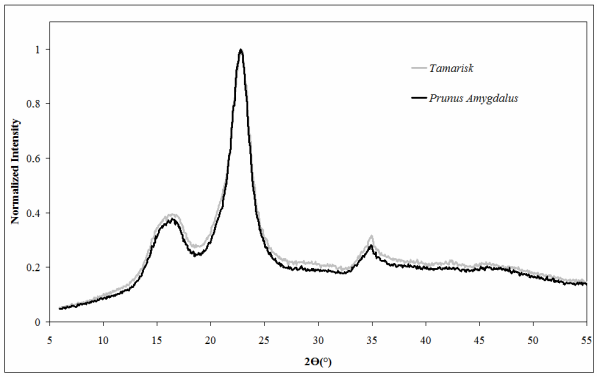


Figure 4: TGA curves of *Prunus amygdalus* and *Tamarisk* sp. fibres

The peak observed in the first region, for both examined fibres, is related to the evaporation of residual water (less than 10% w/w). During the second degradation step (250-450 °C), two degradation mechanisms are observed. The first one (250-350 °C) involves the degradation of hemicelluloses and amorphous cellulose, whereas the second mechanism (350-450 °C) most probably consists in the degradation of the crystalline regions of cellulose. Finally, the last degradation step (T>450) deals with the thermal degradation of lignin. The samples seem to be ash-free pulps.



A)



B)

Figure 5: XRD pattern of *Prunus amygdalus* and *Tamarisk* sp. (A) before and (B) after pulping

XRD analysis of pulps from* Prunus amygdalus *and* Tamarisk *sp.

X-ray diffraction measurements were performed on both extracted fibres, as presented in Figure 5, which shows an intense peak located at a 2θ value of 22.7°, corresponding to the crystallographic plane (002), and related to the crystalline structure of cellulose I for all the samples. Whilst the amorphous background is characterized by a lower intensity diffracted peak at a 2θ value of 18° and attributed to the crystallographic plane (110).[18] According to Segal's method,[18] the crystallinity index of the *Prunus amygdalus* and *Tamarisk* sp. fibre is 72.9 and 70%, respectively. These values are higher than those reported for other annual plants.[14,30,31] The initial raw materials possessed much lower indexes, *i.e.*, 42.6 and 54.6%, for *Prunus amygdalus* and *Tamarisk* sp., respectively. As expected, the crystallinity index increased after pulping, which indicates the removal of amorphous components (hemicelluloses and extractives) and the elimination of lignin sequences.[30,31] However, other peaks were also detected in the case of *Tamarisk* sp. raw material. They correspond to calcium and magnesium elements, as illustrated in the EDX spectra (Fig. 6). These impurities are the most common for lignocellulosic biomass, which can be easily eliminated after cooking and multi-stage washing process, since they are most probably linked with carbonates anions. These findings are in agreement with those deduced from the TGA measurements.

The overall conclusion to be deduced from this part is that despite the difference in the

composition of *Prunus amygdalus* and *Tamarisk* sp. when comparing with other annual biomass,[19,20,32-35] these two residues can be considered as an interesting cellulosic source for papermaking applications. Thus, in the next part, the ensuing fibres were investigated for possible papermaking applications.

Paper characterization

As mentioned in the experimental sections, the prepared fibres were used as such and thus no beating was carried out. The main idea was to valorise a by-product implying the minimum possible operation, in order to stick to a cost-effective approach. The prepared paper sheets were observed by SEM and EDS (Fig. 6). Their physical properties are reported in Table 3.

The SEM micrographs confirmed that the paper from the *Tamarisk* sp. sample was quite homogeneous and resembled those obtained from classical wood fibres (Fig. 6 B). On the other hand, the paper obtained from *Prunus amygdalus* (Fig. 6 A) presented high heterogeneity, which indicated that some uncooked materials were still present (as illustrated in Fig. 6 A), despite the screening and purification treatments.

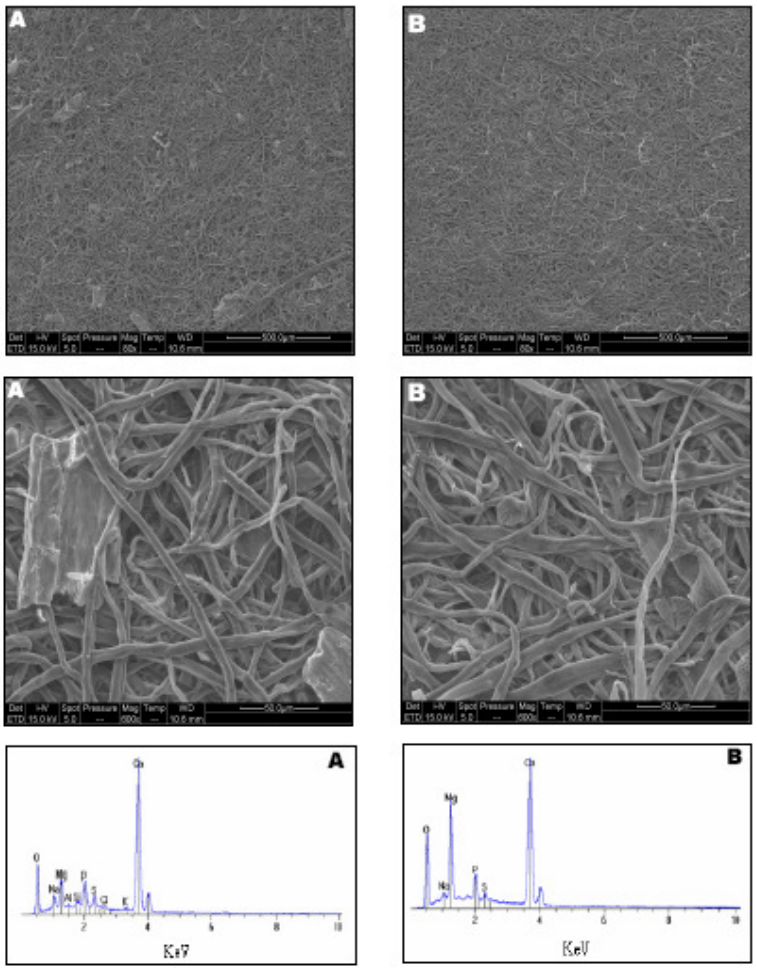


Figure 6: EDS and paper structure of (A) *Prunus amygdalus* and *Tamarisk* sp. (B) using Scanning Electron Micrography (SEM)

The EDS analyses (Fig. 6 C) showed that the predominant residual elements in paper sheets were Ca, O and Mg for *Tamarisk* sp. and Ca and O for *Prunus amygdalus*. The content of these elements was low, if one takes into account that the amount of ashes in these paper sheets was very low. Other minor elements, such as S, P, Na and Al, were also detected.

Concerning the physical properties of the prepared paper sheets, the bulk values (1.72

$cm^3.g^{-1}$ of *Prunus amygdalus* and 1.58 $cm^3.g^{-1}$ of *Tamarisk* sp.) are relatively satisfactory. This value matches very well with that of *Arundo donax* L. reed pulp,[36] but it is lower than those corresponding to *Astragalus armatus*[5] and date palm rachis pulps.[14]

Table 3
Physical properties of paper sheets prepared from *Prunus amygdalus* and *Tamarisk* sp. as well as of those from date palm rachis given for comparison

	Prunus amygdalus	*Tamarisk* sp.	Date palm rachis[14]
Shopper Riegler degree (°SR)	17	21	14
Basis weight (g/m^2)	64 ± 0.8*	64 ± 0.7*	63.9 ± 1.9*
Thickness (µm)	110 ± 8.0*	103 ± 5.3*	141 ± 6*
Bulk (cm^3/g)	1.72	1.58	2.21
Permeabilty ($cm^3/(s.Pa.\ m^2)$)	131.8 ± 0.12*	113.6 ±0.12*	450 ± 0.042*
Breaking length (km)	4.08 ± 0.16*	3.91 ± 0.15*	3.13 ± 0.23*
Elongation, %	0.99 ± 0.08*	0.97 ± 0.08*	1.09 ± 0.09*
Specific energy ($mJ.g^{-1}$)	254.7 ± 22*	239.8 ± 20*	221 ± 37*
Young modulus (GPa)	2.28 ± 0.11*	2.66 ± 0.09*	2.51 ± 0.14*
Burst index ($kPa.m^2.g^{-1}$)	1.38 ± 0.05*	1.0 ± 0.05*	1.32 ± 0.05*
Tear index ($mN.m^2.g^{-1}$)	2.19 ± 0.27*	1.08 ± 0.18*	4.4 ± 0.37*
Dry zero-span breaking length (dry) (km)	13.97 ± 0.85*	9.82 ± 0.70*	13.4 ± 0.91*
Wet zero-span breaking length (wet) (km)	10.65 ± 0.85*	8.69 ± 0.50*	10.8 ± 0.66*
Opacity	99.84 ± 8.5*	99.59 ± 7.8*	94 ± 8.8*

* Standard deviation

The mechanical properties of the paper sheets prepared were evaluated. In fact, the zero-span breaking length values revealed that the intrinsic strength of the fibres was quite high, thus indicating that the cooking conditions were probably suitable. In addition, the other mechanical properties, namely, the values of the breaking length, as deduced from tensile tests (4.08 km for *Prunus amygdalus* and 3.91 km for the *Tamarisk* sp.), Young modulus (2.28 GPa of *Prunus amygdalus* and 2.66 GPa for the *Tamarisk* sp.) and specific energy (254.7 $mJ.g^{-1}$ of *Prunus amygdalus* and 239.8 $mJ.g^{-1}$ for *Tamarisk* sp.), are relatively good for paper handsheets. Indeed, these values are as good as those reported for samples prepared from an unbleached and unbeaten pulp. Moreover, the elongation (1% for both investigated plants), the burst (IB) and the tear (IT) indexes also presented good values (IB was 1.38 and 1.08 $kPa.m^2.g^{-1}$ for *Prunus amygdalus* and the *Tamarisk* sp., respectively, while IT was 2.19 and 1.08 $mN.m^2.g^{-1}$ for *Prunus amygdalus* and *Tamarisk* sp., respectively). These values are in the range of those reported for several other annual biomass,[5,19,20,32-35] but they are lower than those known for particular biomass, such as date palm rachis pulp.[14] Finally, the strength of the fibres (assessed from dry and wet zero-span breaking length) is about reasonably good, *i.e.*, 14 km and 11 km for *Prunus amygdalus* and 10 km and 9 km for *Tamarisk* sp. respectively. These data are particularly promising, if one takes into account that no refining operations were applied before papermaking.

CONCLUSION

The chemical composition of *Prunus amygdalus* and *Tamarisk* sp. was established according to standard methods. The obtained results show clearly that the polysaccharides contents are comparable to those of other annual plants or agricultural crops, which justified their cooking. The isolation of fibres from the starting materials was carried out using soda-anthraquinone and the obtained fibres were characterized by morphological techniques, which confirmed that the two wastes could be very promising sources of cellulose fibres, with a view to using them in several applications, such as composite materials, papermaking, cellulose derivatives etc. Thus, the papermaking valorisation of ensuing pulps gave materials with good properties, without the need of mechanical pre-treatment, *i.e.*, refining operations. This characteristic can be considered as a serious advantage when looking for new alternatives of fibre sources for papermaking.

ACKNOWLEDGEMENTS: The authors express their sincere gratitude to the "PHC-UTIQUE CMCU" (Project No. 13G1114) as well as to the

"Institut Francais de Cooperation en Tunisie-IFC Tunisie, SSHN-2016" for their financial support.

REFERENCES

[1] S. Mansouri, R. Khiari, S. Saadallah and F. Mhenni, *Ind. Crop. Prod.*, **36**, 22 (2012).
[2] A. M. Youssef, M. Ali El- Samahy and M. H. Abdel Rehim, *Carbohyd. Polym.*, **89**, 1027 (2012).
[3] A. M. Youssef, S. Kamel and M. A. El-Samahy, *Carbohyd. Polym.*, **98**, 1166 (2013).
[4] A. A. Hebeish, M. M. Abdelhady and A. M. Youssef, *Carbohyd. Polym.*, **91**, 549 (2013).
[5] Y. Moussaoui, F. Ferhi, E. Elaloui, R. Bensalem and M. N. Belgacem, *BioResources*, **6**, 4969 (2011).
[6] F. Ferhi, S. Das, E. Elaloui, Y. Moussaoui and J. G. Yanez, *Ind. Crop. Prod.*, **61**, 180 (2014).
[7] F. Ferhi, S. Das, Y. Moussaoui, E. Elaloui and J. G. Yanez, *Ind. Crop. Prod.*, **59**, 109 (2014).
[8] A. Omri and M. Benzina, *Desalin. Water Treat.*, **51**, 2317 (2013).
[9] J. Quesada-Medina, P. Olivares-Carrillo and F. J. Lopez-Cremades, *Afinidad*, **68**, 107 (2011).
[10] A. N. Sadegh, M. Kiaei and A. Samariha, *Cellulose Chem. Technol.*, **46**, 369 (2012).
[11] L. P. Xiao, Z. J. Shi, F. Xu and R. C. Sun, *Holzforschung*, **66**, 295 (2012).
[12] L. P. Xiao, Z. J. Shi, F. Xu and R. C. Sun, *Bioenerg. Res.*, **6**, 519 (2013).
[13] M. Ghayeb Zamharir, *Afr. J. Microbiol. Res.*, **5**, 6013 (2011).
[14] R. Khiari, M. F. Mhenni, M. N. Belgacem and E. Mauret, *Bioresour. Technol.*, **101**, 775 (2010).
[15] H. Sihtola, B. Kyrklund, L. Laamanen and I. Palenius, *Pap. Puu*, **45**, 225 (1963).
[16] J. Silvy, G. Romatier and R. Chiodi, *Revue ATIP*, **22**, 31 (1968).
[17] S. Katz, R. P. Beatson and A. M. Scallan, *Svensk Papperstidn.*, **65**, 795 (1984).
[18] L. Segal, J. J. Creely, A. E. J. Mlartin and C. M. Conrad, *Text. Res. J.*, **29**, 786 (1959).
[19] A. Antunes, E. Amaral and M. N. Belgacem, *Ind. Crop. Prod.*, **12**, 85 (2000).
[20] M. N. Belgacem, M. Zid, S. N. Nicolski and A. V. Obolenskaya, *Chim. Technol. Drev.*, *Mej. Sbor. Trud.*, **8**, 111 (1986).
[21] C. H. Chia, S. Zakaria, K. L. Nguyen and M. Abdullah, *Ind. Crop. Prod.*, **28**, 333 (2008).
[22] N. Cordeiro, M. N. Belgacem, I. C. Torres and J. C. V. P. Mourad *Ind. Crop. Prod.*, **19**, 147 (2004).
[23] D. Dutt, J. S. Upadhyaya, R. S. Malik and C. H. Tyagi, *Cellulose Chem. Technol.*, **39**, 115 (2005).
[24] S. Hedjazi, O. Kordsahia, R. Patt, A. J. Latibrai and U. Tschirner, *Ind. Crop. Prod.*, **62**, 142 (2008).
[25] W. D. Wan Rosli, C. P. Leh, Z. Zainuddin and R. Tanaka, *Holzforschung*, **57**, 106 (2003).
[26] D. Dutt, J. S. Upadhyaya, C. H. Tyagi, A. Kumar and M. Lal, *Ind. Crop. Prod.*, **28**, 128 (2008).
[27] J. Gominho, J. Fernandez and H. Pereira, *Ind. Crop. Prod.*, **13**, 1 (2001).
[28] C. Barba, A. De la Rosa, T. Vidal, J. F. Colom, X. Farriol *et al*., *J. Wood Chem. Technol.*, **22**, 249 (2002).
[29] M. T. Goulet and R. A. Stratton, *Nordic Pulp Pap. Res. J.*, **5**, 118 (1990).
[30] P. Saha, S. Manna, S. R. Chowdhury, R. Sen, D. Roy *et al*., *Bioresour. Technol.*, **101**, 3182 (2010).
[31] N. E. Zafeiropoulos and C. A. Baillie, *Composites*: *A*, **38**, 629 (2007).
[32] L. Jimenez and F. Lopez, *Tappi J.*, **73**, 173 (1990).
[33] L. Jiménez, A. Rodriguez, A. Perez, A. Moral and L. Serrano, *Ind. Crop. Prod.*, **28**, 11 (2008).
[34] L. Jiménez, F. Lopez and C. Martınez, *Holzforschung*, **47**, 529 (1993).
[35] L. Ping, P. Brosse Sannigrahi and A. Ragauskas, *Ind. Crop. Prod.*, **33**, 200 (2011).
[36] A. A. Shatalov and H. Pereira, *Bioresour. Technol.*, **96**, 865 (2005).