DYEING OF CHEMICAL PULP WITH NATURAL DYES

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In the present study, plant materials of European flora were investigated as sources of natural dyes in cellulosic fiber dyeing. Specifically, the case of chemical pulp dyeing, using plant materials rich in polyphenolic compounds, such as tannins, anthocyanins and other flavonoids, which are natural colorants, was studied. The most efficient conditions for the aqueous extraction of natural colorants and chemical pulp dyeing process were determined for each plant material, separately. Apart from the final dyeing result, the low cost and minor environmental impact were also taken into consideration during the selection of the most efficient conditions. Therefore, conditions such as short residence time and low temperature were preferred.

Keywords: cellulosic fibers, chemical pulp, dyeing, dyestuff plant materials, extraction, natural colorants

INTRODUCTION

Natural dyes are generally organic compounds that are soluble in a solvent, usually in water, and come from diverse places, such as the roots of a plant, the bark of a tree, a parasitic insect and even lichens.[1,2]Nevertheless, since the beginning of the 20th century, dyes are produced almost exclusively from the by-products of the petrochemical industry. This fact is based on the invention of the synthetic organic chemistry and the desire for a great variety of bright and stable colorants.[3]

Nowadays, there is a worldwide resurgence of interest in natural dyes, in the context of the general return to natural resources. This trend is based firstly on the fossil fuel depletion and secondly on the increasing public awareness of the environmental and health hazards caused by chemical products in general and, subsequently, by synthetic dyes.[4-8] The syntheses and applications of synthetic dyes may have adverse effects on the health of the sensitive groups of population, such as children and asthmatic or allergic persons, due to their allergenicproperties.[5]

Moreover, natural dyes which are derived from wastes, such as the coloured plant wastes released from the food and beverage industry, agricultural activities and forestry, such as vegetable and fruit peels or timber residues, and non-edible plants, such as weeds, show vital importance related to sustainability.[4,5,9]

The process of colouring fibrous products is very complex, but it is based on the simple mechanism of binding a chromophore,a chemical compound that brings colour, to the surface of the fibers, or of trapping it within them.[10,11]Natural colorants could be classified intofive groups, the polyphenolic compounds, such as flavonoids and tannins, the terpenoids, the tetrapyrroles, the quinines and the N-heterocyclic compounds.[1,2]

At this point, it is important to refer to the disadvantages of using natural dyes. The most important are their low resistance against light and water and the difficulty of achieving a standard dyeing result. Natural dyes, when used by themselves, have many limitations of fastness and brilliancy of shade. However, when used along with metallic mordants, they produce bright and fast colours. The function of the mordant is to assist the adsorption of the dye and promote good bonding of dye and fiber as a bridge, which helps to bond fiber and natural dyes at the molecular level.[7,12-14]The characteristic property of natural dyes to show limited fastness and resistance against leaching, when they are used without any mordant addition, represents a key factor in the paper recyclingprocess.[1,14]For this reason, their application in paper industry is of great importance.

Chemical pulp is a fiber of cellulose $(C_6H_{10}O_5)_n$, which is obtained from plant material (95% from wood) and is further processed predominantly in the paper industry. Chemical pulping achieves the isolation of cellulose fibers from the treated plant material by degrading the lignin and hemicelluloses into small, water-soluble molecules, which can be washed away from the cellulose fibers avoiding their depolymerization. The kraft process (mixture of sodium hydroxide and sodium sulfide) is the dominant chemical pulping method, the sulfite process being the second; while soda pulping is the first successful chemical pulping method.[15]The optimization of chemical pulp dyeing using natural sources is crucial, considering that it represents almost 70% of the worldwide wood pulp produced annually. In addition, chemical pulp lacks lignin in its composition, consequently larger amounts of dye are required for achieving adequate dyeing results, in comparison with mechanical pulp, which consists of up to 20% w/w of lignin.[15-17]

Finally, according to recent literature, natural dyes show significant antimicrobial activity, imparting unique properties to the dyed natural fibers. The plants synthesize aromatic substances, such as alkaloids, terpenoids, and phenolic compounds as their secondary metabolites. These chromophore compounds are antimicrobial and are produced by the plants in response to an attack by a pathogen.[18,19] Therefore, the use of these chromophore compounds in pulp dyeing can lead to paper products with antimicrobial activity, which is very interesting in the case of functional food packaging, cleaning papers and paper products for personal hygiene. The dyeing of mechanical and chemical pulp with natural dyes is a research area of great interest and thoroughly investigated by our research group.[20,21]

Therefore, in the framework of this study, experiments were performed to evaluate the effect of extraction and dyeing process conditions on the dyeing capacity of various dyestuff plant materials on chemical pulp. The estimation of the dyeing capacity occurred through the determination of colour coordinates of the dyed paper, according to the CIE $L^*a^*b^*$ system.The natural colorants used were extracted from four different plant species that are found in abundance in European flora and are rich in polyphenolic compounds, such as tannins, anthocyanins and other flavonoids, which are natural colorants. Specifically, the dyeing capacity of red onion (*Allium cepa* L.) peels, eucalyptus (*Eucalyptus globulus* L.) bark, inula (*Inula graveolens* L.) leaves and quince tree (*Cydonia oblonga* Mill.) leaves on chemical pulp was examined. In Table 1, the main characteristics and colour constituents of each dyestuff plant material are presented.

EXPERIMENTAL

Production of the chemical pulp

The origin of the chemical pulp used was spruce softwood with moisture content of 7% w/w, supplied by the Athenian Paper Mills – Softex. According to the supplier, the chemical pulp was bleached pulp obtained through alkaline Kraft delignification of spruce tree trunk, using a mixture of sodium hydroxide and sodium sulfide, the latter beingproduced in the recovery process by the reduction of sodium sulfate.The supplied chemical pulp was treated according to the following procedure. Firstly, it was torn into small pieces by hand and added into deionized water in a plastic pot, so that the resulting pulp stock consistency was 2% w/w. Then, vigorous mixing of the resulting mixture took place for 6 h, using a mechanical mixer at 2000 rpm. After mixing, in order to reduce the retained water, the produced pulp was filtered through a Buchner funnel. After the filtration, the moisture content of the pulp was 92% w/w. At the end, the produced pulp was stored in an airtight plastic pot at 4 ^oC.

Selection and storage of plant material

The selection of the dyestuff plant materials was based on the sustainability and, more so,on the economic and environmental efficiency of the production of natural dyes. Therefore, the use of non-edible dyestuff materials derived particularly from wastes and by-products of several procedures, such as agricultural activities and forestry, are recommended. In this case, the supply cost of the dyestuff material is low, even zero, the use of cultivated land for non-edible crops is avoided and, moreover, this is an alternative and environmentally friendly way of waste disposal. Specifically, *Allium cepa* L. peels and *Cydonia oblonga* Mill. leaves are agricultural residues that occur in great quantities after crop harvesting and remain on the field. Additionally, eucalyptus wood is widely used in paper-making industry because of its rapid growth rate and demand for minimal engineering changes in pulp mills. However, in order to obtain quality paper, a debarking treatment of eucalyptus wood is needed. Thereby, about 15-20% of the eucalyptus wood utilized represents bark. At an estimated amount of 2.5 million tonnes of eucalyptus wood used per annum, about 0.3-0.5 million tonnes of bark is generated.[22]Therefore, an alternative use of eucalyptus bark, as a source of natural dyes to be used in pulp dyeing, is crucial for paper-making industries. Finally, *Inula graveolens* L. is a widely known herb in Mediterranean flora, which is often found as a weed in rural cultures and thrives in temperate climates.

The selected plant materials were naturally dried in a dark place at room temperature (25 ^{o}C) and relative humidity (RH) of approximately 50%, for one week. The dried dyestuff plant materials were ground into pieces and stored in a dark and dry place. In Table 1, the used plant materials and their main characteristics are presented.

Extraction

For the production of natural dyes, aqueous extraction of the selected plants was carried out, as it is a mild and non-polluting procedure, which permits the further exploitation of the solid residues as animal feed or soil conditioner.

The examined extraction conditions varied from 40 to 80 ^{o}C extraction temperature (T_e) and from 15 to 60 min residence time (t_e), all the possible combinations were examined. Every time, specific amounts of the ground plant material were extracted with 125 ml of deionized water. In each experiment, the specific amounts of extracted plant material were determined from the Φ/X ratio. This ratio refers to the oven dry weight of the extracted plant material to the oven dry weight of the pulp dyed with the plant extract. The final extract was obtained after filtration of the insoluble residues through a copper filter fabric.

Dyeing

Each produced plant extract was used for the dyeing of chemical pulp. Dyeing was realised using a liquor ratio of 55:1 mL/g (for 1.8 g of oven-dried pulp, a dyebath of 100 mL was used) at various combinations of temperature (T_d) and residence time (t_d) (from 25 to 80 ^{o}C and from 15 to 60 min, respectively). All the dyeing experiments were performed in triplicate and one sheet was formed from the dyed pulp of each experiment.

Table 1
Characteristics of the used dyestuff plant materials

Plant	Used part	Grain size (mm)	Moisture content (% w/w) of naturally dried material	Main dyestuff components
Allium cepa L.	Peels	5.00-10.00	11.00	Anthocyanins and flavonoids: quercetin, rhamnetin[23]
Cydonia oblonga Mill.	Leaves	4.00-5.00	7.60	Tannins, flavonoids and carotenoids[24]
Eucalyptus globulus L.	Bark	1.40-1.00	8.07	Tannins and flavonoids: eriodictyol, naringenin, quercetin, rhamnazin, rhamnetin, taxifolin[25]
Inula graveolens L.	Leaves	0.50-1.00	10.25	Flavonoids: sakuranetin, kaemperol[26]

Sheet formation

Sheet formation from the dyed and undyed pulps was performed according to SCAN-C-26-76 and SCAN-C-M5-76 methods, using a Lorentzen and Wettre (Sweden) sheet former, model SCA, a laboratory sheet press performing at 2.5 MPa for 4 min and a Lorentzen and Wettre rapid dryer, where the wet sheets were dried for 17 s at 180 ^{o}C.

The produced sheets had a diameter of 165 mm, oven dry mass of 1.8 ± 0.05 g and were stored in a dark and dry place for at least 24 h before their colour determination.

Colour measurement

The colour of the produced sheets was determined according to the CIE $L^*a^*b^*$ system. The chromatic coordinates, $L^*a^*b^*$, were measured using a Dr. Lange colourimeter. The CIELAB colour space is organized in a cube form. The vertical L^* axis represents lightness and runs from top to bottom, while the horizontal axes are represented by chromatic coordinates a^* and b^*. The chromatic coordinate a^* represents the difference between green ($-a^*$) and red ($+a^*$), and b^* represents the difference between yellow ($+b^*$) and blue ($-b^*$). If the CIELAB colour space is cut into halves, a coloured circle is seen. Around the edge of the circle, every possible saturated colour, orhue is presented. This circular axis is known as h° for Hue. The units are degrees° (or angles), ranging from 0° (red) through 90° (yellow), 180° (green), 270° (blue) and back to 0°.The Hue is given from the following equation:

$$h = tan^{-1}\left(\frac{b^*}{a^*}\right) \quad (1)$$

The colour measurements were carried out on the upper side of the dyed sheets in three different spots creating an imaginary triangle. As final value for the chromatic coordinates of each sheet, we chose the average value of the afore-mentioned three measurements. The final dyeing result was determined from the total colour difference ΔE^*_{ab} between each dyed sheet and an undyed sheet called reference sample. The higher the ΔE^*_{ab}, the more efficient the dyeing result is, as the pulp fibers show better absorption of the dye. It is noteworthy that each time the dyed sheets and the reference sample should come from the same chemical pulp. The total colour difference is given from the following equation:

$$\Delta E^*_{ab} = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (2)$$

where $\Delta L^* = L^*_{sample} - L^*_{reference}$, $\Delta a^* = a^*_{sample} - a^*_{reference}$ and $\Delta b^* = b^*_{sample} - b^*_{reference}$

Moreover, the percent reflectance values (at λ_{max} = 500 nm) were recorded and colour strength values (K/S) were calculated according to the equation:

$$\frac{K}{S} = \frac{[1 - 0.01R]^2}{2 * [0.01R]} \quad (3)$$

where R: spectral reflectance of the sample in %, K: absorption and S: scattering.

The Kubelka-Monk equation is useful when formulating colors for industries, such as textiles, paper, and coatings. For these applications, it is assumed that the scattering (S) of a dye or pigment depends on the properties of the substrate or opacifier, while the absorption (K) of light depends on the properties of the colorant.

RESULTS AND DISCUSSION

In the present work, the optimization of dyeing and extraction conditions (T_d, t_d, T_e and t_e, respectively) was based on the best combination of dyeing result, cost and environmental impact.

Optimization of dyeing conditions

Savvidou M. and Economides D. showed that extraction conditions around T_e = 80°C and t_e = 60 min are severe and give extracts with high content of colouring agents.[21] Moreover, a residence time of 30 min in the dyebath (t_d) presents strong dyeing results.[20] Therefore, in order to determine the most efficient dyeing temperature (T_d), the above conditions remained constant and T_d of 25, 40 and 80 °C was examined. In Fig.1a-d, the relation of the Φ/X ratio and ΔE^*_{ab} for each plant is presented.

Firstly, Fig.1a-d shows that there is a specific limit of the Φ/X ratio, which is characteristic of each plant material and depends on extraction and dyeing methods. Beyond this limit, any increase of the Φ/X ratio is unable to cause any further important colour change, as the pulp fibers cannot take up any more dye. Therefore, Savvidou M. and Economides D. named this limit "the limiting saturation value".[21] The determination of the limiting saturation value (Φ/X) is a fact of particular importance as it constitutes a decisive factor for the optimization of the chemical pulp dyeing process. The limiting saturation value is different for each dyestuff plant material and moreover, it gives the possibility to forecast the Φ/X ratio where it is possible to achieve the tensest dyeing result. This forecast leads to important saving of resources and energy, avoiding the pointless consumption of additional quantities of dyestuff plant material, while beyond the limiting saturation value, the total colour difference remains relatively immutable. The limiting saturation value for the afore-mentioned extraction and dyeing conditions is presented in Table 2.

Furthermore, it may be concluded from Fig.1 that *Eucalyptus globulus* L. bark gives high ΔE^*_{ab} at all the examined dyeing temperatures T_d, even at the lowest. In this case, the room temperature is the most efficient dyeing temperature, as long as the dyeing result is satisfactory, and for these conditions the cost is low. It should be noticed that according to the literature, when the dyeing result is tense, the human eye cannot realize the slightly different shades, unless the ΔE^*_{ab} difference between the samples under examination is above 4 units.[27]

Table 2
Limiting saturation value for each plant (T_e= 80°C, t_e= 60 min, T_d optimum, t_d= 30 min)

Plant material	T_d (°C)	Limiting saturation value	ΔE^*_{ab}
Eucalyptus globulus L.	25	1.4	27.0
Allium cepa L.	40	1.4	29.9
Inula graveolens L.	40	1.8	10.9
Cydonia oblonga Mill.	25	2.2	11.1

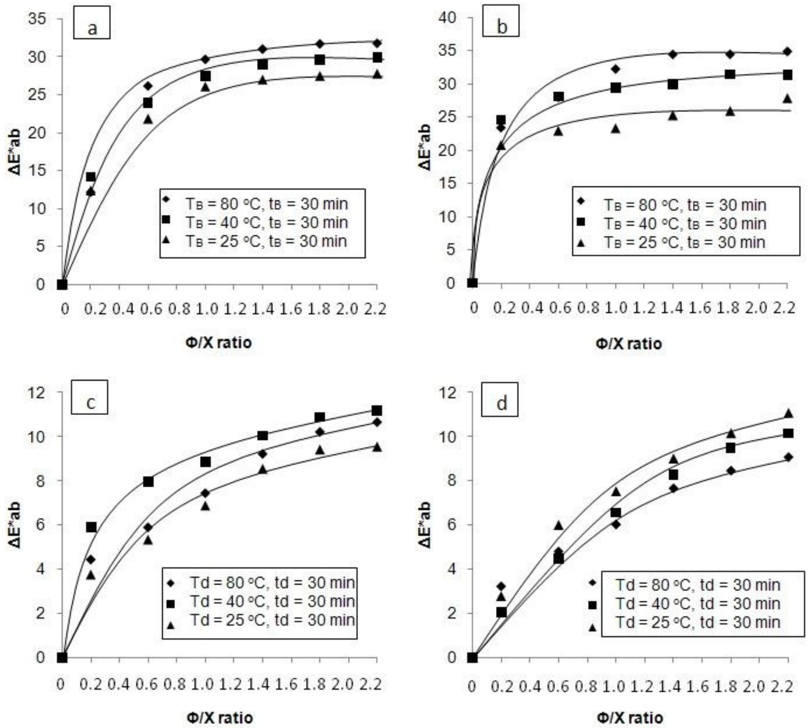

Figure 1: Impact of dyeing temperature on $\Delta E^*_{ab}$and its relation with the Φ/X ratio (T_e= 80 °C, t_e= 60 min, t_d= 30 min); a. *Eucalyptus globulus* L. bark, b. *Allium cepa* L. peels, c. *Inula graveolens* L., d. *Cydonia oblonga* Mill.

Table 3
Impact of residence time in the dye bath on total colour difference of the sheets produced from chemical pulp dyed with plant extracts (T_e= 80°C, t_e= 60 min, T_d optimum)

	Total colour difference ΔE^*_{ab}											
	Eucalyptus globulus L. T_d = 25°C			*Allium cepa* L. T_d = 40°C			*Inula graveolens* L. T_d = 40°C			*Cydonia oblonga* Mill. T_d = 25°C		
t_d (min) / Φ/X ratio	15	30	60	15	30	60	15	30	60	15	30	60
0	0	0	0	0	0	0	0	0	0	0	0	0
0.2	10.6	12.4	12.6	18.0	24.6	24.5	3.9	5.9	5.0	1.9	2.8	3.1
0.6	16.9	21.8	22.9	21.4	28.1	27.2	5.2	8.0	6.6	5.0	6.0	7.1
1.0	20.8	26.1	25.6	26.9	29.4	31.1	6.7	8.8	8.4	6.9	7.5	8.1
1.4	23.0	27.0	27.0	29.1	29.9	30.9	7.9	10.1	9.2	7.4	9.0	10.3
1.8	23.9	27.4	28.0	30.9	31.5	32.5	8.0	10.9	9.9	8.2	10.2	11.2
2.2	24.1	27.8	28.1	30.8	31.4	31.8	8.9	11.2	10.8	9.0	11.1	11.6

So, a dyeing temperature of 25 °C is preferred to that of 80 °C, despite the fact that the latter

Similarly, taking into consideration the optimum combination of tense dyeing result, cost effectiveness and eco-friendliness, the preferred $T_d$for *Allium cepa* L. peels and *Inula graveolens* L. is 40°C and for *Cydonia oblonga* Mill. leaves is 25 °C.

shows a slightly stronger dyeing result (ΔE^*_{ab} difference ≈ 4 units at saturation limit).

Since the optimum $T_d$had been determined for each plant material, the optimization of t_d was investigated. Therefore, a t_d range from 15 to 60 min was examined. In Table 3, the relation between the residence time in the dye bath and total colour difference is presented for each plant material, keeping constant all the other parameters (T_e= 80°C, t_e= 60 min, T_d optimum). According to Table 3, the optimum t_d for *Eucalyptus globulus* L. bark, *Allium cepa* L. peels and *Inula graveolens* L. is 30 min, as this residence time is relatively short and gave an equal dyeing result with t_d = 60 min. For a shorter residence time, the dyeing result is very weak in comparison with 30 and 60 min, especially at low Φ/X ratios. In the case of *Cydonia oblonga* Mill., the optimum t_d is 60 min, as the dyeing result is weak even with high Φ/X ratios, therefore every difference in ΔE^*_{ab}, even the slightest, is significant.

Optimization of extraction conditions

The optimum extraction conditions were determined for each plant material keeping constant the optimum dyeing conditions as they are mentioned above. Moreover, the examined extracts were obtained at an Φ/X ratio of 0.6 units in order to avoid the saturation limit, and every colour change would be significant. Extraction temperatures of 40 and 80 °C for 15, 30 and 60 min residence time in all possible combinations were examined. In Fig.2a-d, the impact of the extraction conditions (T_e, t_e) on total colour difference (ΔE^*_{ab}) for each plant is presented. Additionally, in Table 4, the chromatic coordinates of the sheets produced from the chemical pulp dyed with plant extracts under optimum dyeing conditions for Φ/X = 0.6 are exhibited, in order to justify the impact of the different extraction conditions on the final dyeing result.

According to Fig.2a-d, the optimum extraction temperature was determined as 80 °C, in all studied cases. Lower T_e gave weak final dyeing results (Fig.2a-d, Table 4). The optimum extraction residence time for *Eucalyptus globulus* L. bark and *Allium cepa* L. peels was 15 min and 60 min in the case of *Inula graveolens* L. and *Cydonia oblonga* Mill. leaves (Fig.2a-d). Furthermore, through the chromatic coordinates (Table 4), the achieved hues could be determined. Specifically, *Eucalyptus globulus* L. gave strong red-brownish hues, *Allium cepa* L. yellow ones, *Inula graveolens* L. bright yellow-green ones and *Cydonia oblonga* Mill. bright red-yellowish ones.

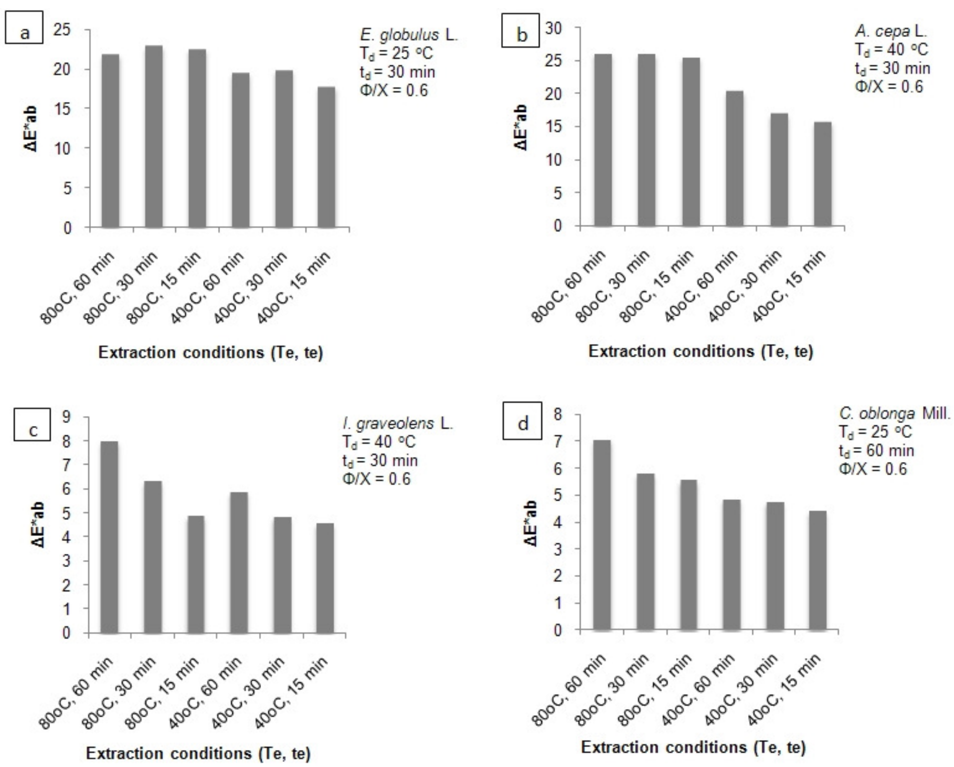

Figure 2: Impact of extraction conditions on ΔE^*_{ab}; a. *Eucalyptus globulus* L. bark, b. *Allium cepa* L. peels, c. *Inula graveolens* L., d. *Cydonia oblonga* Mill.

Table 4
Impact of different extraction conditions on chromatic coordinates of the sheets produced from the chemical pulp dyed with plant extracts under optimum dyeing conditions for Φ/X = 0.6

Extraction conditions (T_e, t_e)	Chromatic coordinates, $L^*a^*b^*$																			
	Eucalyptus globulus L.					*Allium cepa* L.					*Inula graveolens* L.					*Cydonia oblonga* Mill.				
	L^*	a^*	b^*	h	ΔE^*_{ab}	L^*	a^*	b^*	h	ΔE^*_{ab}	L^*	a^*	b^*	h	ΔE^*_{ab}	L^*	a^*	b^*	h	ΔE^*_{ab}
40°C, 15 min	82.8	5.9	11.1	62.0	17.8	90.8	0.3	15.2	88.9	15.8	96.2	-1.6	5.1	107.4	4.5	94.9	0.5	4.7	83.9	4.4
40°C, 30 min	80.7	6.9	10.3	56.2	19.9	87.2	1.3	17.1	85.7	17.0	95.3	-1.6	4.9	108.1	4.8	94.7	0.7	5.0	82.0	4.7
40°C, 60 min	81.5	7.5	10.3	53.9	19.5	86.2	2.0	18.3	83.8	20.4	95.1	-1.5	6.1	103.8	5.9	94.4	0.9	5.0	79.8	4.8
80°C, 15 min	79.0	8.2	13.2	58.2	22.5	84.1	-0.2	23.2	90.5	25.6	95.6	-1.6	5.1	107.4	4.8	94.3	0.9	5.7	81.0	5.5
80°C, 30 min	79.0	8.6	13.6	57.7	22.9	84,1	-0,1	23,3	90.2	26.0	94.9	-1.6	6.4	104.0	6.3	94.0	1.1	5.9	79.4	5.8
80°C, 60 min	78.8	8.1	12.9	57.9	21.8	81.8	1.4	22.6	86.5	26.1	93.3	-1.5	7.4	101.5	8.0	93.1	1.8	6.8	75.2	7.0

Table 5
K/S values at λ_{max} = 500 nm for optimum dyeing results of each dyestuff plant material at Φ/X = 0.6

Plant material	K/S	ΔE^*_{ab}
Eucalyptus globulus L.	0.28	22.5
Allium cepa L.	0.15	25.6
Inula graveolens L.	0.02	8.0
Cydonia oblonga Mill.	0.04	7.0

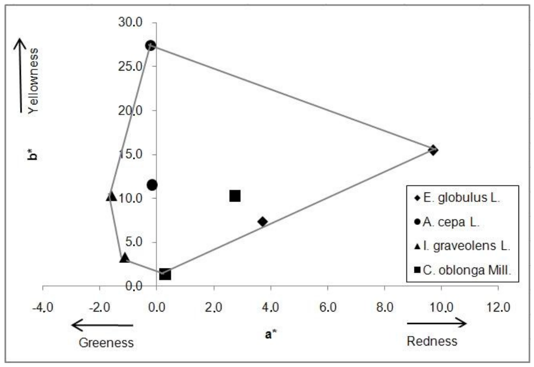

Figure 3: Plot showing the colour gamut achieved by the application of four different plant extracts on chemical pulp

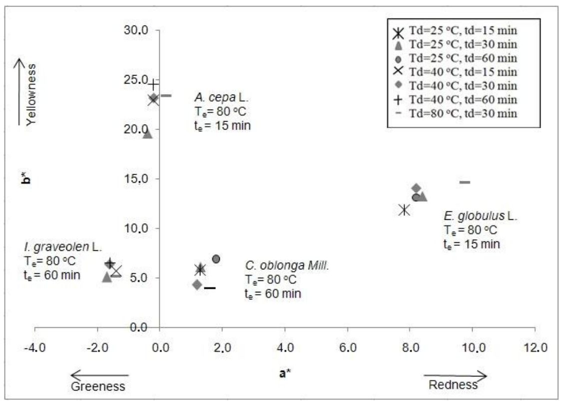

Figure 4: Plot showing the impact of dyeing conditions (T_d, t_d) on colour shade at Φ/X ratio = 0.6

In Table 5, the K/S values at λ_{max} = 500 nm are presented for selected paper sheets dyed under the optimum dyeing conditions, using the plant extracts resulted from the optimum extraction conditions, for each dyestuff plant material (see Figs. 1 and 2 and Tables 3 and 4).

In Fig.3, both the strongest and the weakest dyeing result of each plant material on dyed chemical pulp is presented. To ensure the strongest shade, experiments were performed at an Φ/X ratio of 2.2 units (beyond the saturation limit) under optimum extraction and dyeing conditions. Additionally, to ensure the weakest shade, experiments were performed at an Φ/X ratio of 0.2 units under less effective extraction and dyeing conditions. In both cases, the examined extraction and dyeing conditions arise from the previous analysis. The area of the a^*b^* plot that is delineated from the experimental points, which resulted for pure plant extracts, represents the colour gamut that could be achieved by mixing the pure extracts. So, from Fig.3, it may be noted that a wide range of yellow-red shades could be achieved from the extracts of the examined plant materials.

Finally, the impact of dyeing conditions (T_d, t_d) on the achieved colour shade is presented on the a^*b^* plot (Fig. 4). Therefore, the occurring colour shades were examined firstly for constant t_d=30 min at T_d= 25, 40, 80^{o}C and subsequently, at constantT_d (the optimum for each plant material) for t_d= 15, 30, 60 min. In all cases, the used extracts were produced under the optimum extraction conditions for each plant and anΦ/Xratio of 0.6 units. From Fig. 4, it is assumed that the colour shade in all cases depends primarily on T_d, while the impact of t_d is not significant.

CONCLUSION

The presented results prove that dye stuff plant materials, especially from agricultural and forestry wastes, could be used sufficiently as sources of natural colorants in the dyeing process of chemical pulp. In this case, an alternative application of natural dyes is revealed beyond the usually recommended applications such as textile dyeing, food and cosmetics colouring. Pulp dyeing is an industrial activity of great importance due to the extremely wide range of products made from dyed pulp, such as packaging materials, printing papers, household care and personal hygiene papers.

The application of natural dyes on chemical pulp hides some drawbacks, such as the inability of reproducing the dyeing result and its dependence on the extraction conditions primarily and on dyeing conditions secondly, which vary for each plant source, and finally the high cost of these procedures. In this study, these important weaknesses were overcome by determining the most efficient extraction and dyeing conditions for each plant, according to the best combination of dyeing result, cost and environmental impact. Therefore, the selected conditions not only led to efficient dyeing, but also required minimum temperature and short residence time.

Finally, it was determined that in order to achieve a sufficient dyeing result, approximately the same amount of plant material and pulp mass is needed. According to the presented experimental results, the further addition of plant material leads to the saturation point, where the pulp fiber is unable to take up more dye. The achieved shades occurred from the selected dyestuff plant materials cover the yellow – red colour area in the CIELAB colour space, by achieving hues from light yellow to reddish brown.

For further study, the use of other technologies, such as the ultrasound and microwave, in the extraction and dyeing procedure is suggested. In this case, the achievement of a very tense dyeing result is expected even at lower temperature, shorter residence time and lower Φ/X ratio.

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