FTIR Spectroscopic, Colorimetric and Statistic Evaluations of Textile Materials Dyed with Crude Extract Obtained by Macerating Pomegranate Peels

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Crude extract obtained by macerating pomegranate peels can dye textile materials. The final purpose of the process of pomegranate peels maceration in the presence of glacial acetic acid was to extract pigments that can dye various textile materials: wool, polyamide, cotton and cellulose acetate. Dyeing of these materials started with pre-mordanting with citric acid (for 15 min) and continued with dyeing in itself at 100°C for 60 min. The crude extract obtained at the end of the process of pomegranate peel maceration. The acid pH of the dyeing liquor generated by the presence of acetic acid, unseparated from the extracted pigments constituted an advantage for wool and polyamide dyeing, but it was not favorable for cotton and cellulose acetate dyeing. The pigments presence on the dyed textile materials has been proved by FT-IR spectroscopic analysis. Quantitative evaluations consisted in colorimetric measurements and statistic analysis of textile materials dyeing capacity (using the total difference of color, ΔE^*).

Keywords: crude extract, peel pomegranate, FT-IR, color measurements, statistical analysis

The color of textile materials acquired by dyeing with pigments extracted from pomegranate peel depends on the quality, structure and color of these natural dyes. Pomegranate peel has a complex composition consisting of polyphenols, tannis, flavonoids and complex polysaccharides. The pigments present in the peel of Punica granatum are called anthocyanidins. The presence of anthocyanidins in pomegranate peel was proved by chromatographic and UV-Vis analyses [1-4].

The anthocyanidins from pomegranate peel are: Pelargonidin (Pg), Cyanidins (Cy) and Delphinidin (Dp). These pigments are water-soluble compounds and provide red-blue, orange and violet colors. Figure 1 presents the structure of these anthocyanidins and the colors generated by them.

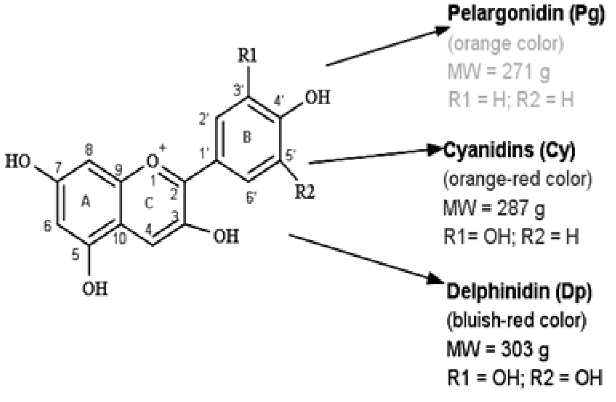

Fig. 1. Chemical structure of anthocyanidins from pomegrate peel [5]

Anthocyanidins (or aglycons) consist of 3 aromatic rings, A, B and C. The aromatic ring A is bound to a B heterocycle (which has 5 atoms of C and one atom of O) which, in turn, is bound to the aromatic ring C, by means of a C-C bond, in ortho-position related to oxygen [6]. If at the level of A and B rings in the positions 3 and 5, there are glucosidic rings, these compounds are called anthocyanins. The color stability of these natural pigments depends on *p*H, temperature, solvent, light, the presence of other compounds (ex. Flavonoids, phenolic acid and metals) and chemical structure [7, 8]. Anthocyanidins chemical structure depends on the following factors: *p*H, the structure of the chromophore group, the presence and number of OH groups on the aromatic B ring, methylation degree of these OH groups, number of aliphatic/aromatic groups attached to sugar, number and position of sugars attached [8].

Red chromophore from anthocyanidins can be found very rarely in plants in free state and only as polyhydroxylated salts.

Anthocyanidins color can vary from red to blue under the *p*H influence, as follows:

-at *p*H = 1, the orange to red flavylium cation is formed in anthocyanidins;

-at *p*H = 2÷4, blue colored quinoidal species are formed in anthocyanidins;

-at *p*H = 4.5 anthocyanidins become colorless, because the carbinol pseudobase (hemiketal form) through nucleophilic attack of the position 2 of flavylium cation, where an OH group will be attached;

-at *p*H ≥7, anthocyanidins's color is blue due to the formation of quinoidal base.

Knowing these aspects can be useful in the realization of the experimental protocol of textile materials dyeing process (for a correct choice of dyeing bath pH, depending on the type of textile materials subjected to dyeing, and the natural pigments color).

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			Sample codes			
Crt. No.	Treatment code	M : CE[a] (mL:mL)	Wool	Polyamide	Cotton	Cellulose mono-acetate
1	Witness	Witness	0:0 W	0:0 PA	0:0 C	0:0 CMA
2	0CA	0:12	0:12W	0:12PA	0:12C	0:12 CMA
3	1CA	6:6	6:6W	6:6PA	6:6C	6:6 CMA
4	2CA	3:9	3:9W	3:9PA	3:9C	3:9 CMA
5	3CA	9:3	9:3W	39:3PA	9:3C	9:3 CMA
6	4CA	5:14	5:14W	5:14PA	5:14C	5:14 CMA

Table 1
EXPERIMENTAL PROTOCOL

[a] M : CE is the volumetric ratio for mordant: crude extract

Dyeing the textile materials (cotton and synthetic) with dyes extracted from pomegranate peel or arils by means of a solvent (ethanol, methanol, water) is a less studied topic, due to the large number of components and the complexity of the chemical structure of the pigments existing in the peel, juice or arils [9-11].

Literature also presents some studies concerning wool and natural silk dyeing with crude dye and dye extracted from pomegranate peel, pigments obtained by using ethanol as solvent [12, 13].

This paper presents the dyeing process for 4 types of textile materials (wool, polyamide, cotton and cellulose acetate) by means of crude extract obtained by pomegranate peel maceration for 6 months. The solvent used in the solid-liquid extraction process was concentrated acetic acid, which provided an acid *p*H (*p*H ≤ 4) and in which the natural pomegranate pigments have colors varying from red-orange to blue. One has used the dyeing technology based on pre-mordanting with citric acid for 15 min. The IR spectra and the quantitative analyses (color measurements and statistic analysis) confirmed the dyeing.

Experimental part

Materials and methods

Textile materials were acquired from Romanian textile companies, as follows: wool from Carpatex Brasov, and other textile materials from Iasitex, Iasi. The acetic acid (the solvent) was obtained from Merck Co.

Solid-liquid extraction: maceration of pomegranate peel (50 g) lasted 6 months and was performed by means of glacial acetic acid (100 mL). The obtained extract was used in its crude form, without a previous distillation to separate the solvent (acetic acid).

Textile materials dyeing was performed in two stages: 1) mordanting with citric acid for 15 min; 2) dyeing in itself with crude extract obtained at the end of the solid-liquid extraction process. Table 1 presents the experimental protocol and the 5 volumetric mordant: extract ratios. The total volume of each dyeing liquor was of 120 mL; the four types of textile materials (cut in 10x10 cm samples) were present in the dyeing vessel each time. Each dyeing operation was repeated 10 times, to enable a statistic analysis of the capacity of textile materials dyeing with crude extract obtained from pomegranate peels. The dyeing was performed on Mathies Policolor Machine at 100°C during 60 min. Intense washing in hot and finally cold water followed the dyeing.

Methods of analysis

FT-IR – analysis was performed on a Spectrophotometer FTIR IRAffinity-1 Shimadzu (Japan). The spectra registration was realized with 250 scans in the 4000–600 cm^{-1} range. The overlapping spectra were performed using Jascow32 software.

Colour strength (K/S), *CIE L*a*b* measurements (L*, a*, b*, C*, h*) and CIE L*a*b* color differences (ΔE*, ΔL*, ΔC*, Δa*, Δb* and Δh*)* were measured on a Datacolor Spectrophotometer Spectroflash SF300. The significances of the color *measurements* [14-26] are as follow: L* is the lightness/luminosity; a* and b* indicate the red-green and yellow-blue coordinates; C* is the saturation of color and h* is the hue of color. The correspondent color differences are: ΔE* - the total differences of color; ΔL* - the difference in colour between the examined sample and witness, i.e. the distance between the respective positions in CIE LAB space; ΔL* - the difference in lightness; ΔC* - saturation difference; Δa* and Δb* are the differences on the red-green coordinate respective on the blue-yellow coordinate; Δh* - the hue difference.

Statistical analysis: the dyeing capacity of samples were tested using statistical analysis of the total differences of color values for dyed samples. The dyeing capacity testing was performed determining the total differences of color (ΔE*) for 10 dyed samples realized in the same conditions, for each material type. These values were then used to calculate the standard deviation (SD), the standard error of the mean (SEM) and the coefficient of variation (CV). These values were calculated using a MATLAB software [27, 28] where the standard deviation SD of the set of distinctive statistic data $\{(\Delta E^*)_1, (\Delta E^*)_2, \ldots(\Delta E^*)_{10}\}$ was calculated with the eq. (1) [29]:

$$SD = \left[\frac{\sum_{i=1}^{n} \left((\Delta E^*)_i - \overline{(\Delta E^*)} \right)^2}{n} \right]^{1/2} \qquad (1)$$

where:

SD - standard deviation (shows the total differences of color values dispersion);

n - total number of observations (i = 1-10, resulting n = 10).

The standard error of the mean and the coefficient of variation were calculated using the eqs. (2) and (3), as follow:

$$SEM = \frac{SD}{n^{1/2}} \qquad (2)$$

where:

SEM - standard error of the mean;

n - number of observations (n = 10).

$$CV = \frac{SD}{\overline{(\Delta E^*)}} \times 100 \qquad (3)$$

where:

CV - coefficient of variation;

SD - standard deviation;

$\overline{(\Delta E^*)}$ - the average of total differences of color.

If the variation coefficient (CV) of ΔE* values is lower, then the dyed samples have a similar dyeing capacity and indirectly, results that the uniformity of the dyed samples is better.

Results and discussions

The color of the textile materials dyed with crude extract obtained from pomegranate peels is given by the color of tannins and pigments extracted during mordanting process.

The color of crude extract obtained from pomegranate peels by means of concentrated acetic acid is brown and it is given by the combination of the colors of the three extracted anthocyanidins present in the form of flavlium or quinoidal species: orange (from Pg) + bluish-red (from Dp) + orange-red (from Cy) = brown. The color of tannins extracted from pomegranate peel is also brown.

All the dyeing operations performed during this study were carried out at an acid *p*H, ensured by the presence of solvent/concentrated acetic acid, and that of the mordant (citric acid). This means that the anthocyanidins from the crude extract have colors ranging from red-orange to blue; therefore, they preserve their color.

Color strength, K/S

The volumetric mordant/crude extract ratio influences color strength values, K/S. One can notice in figure 2 that the brown extract obtained from pomegranate peels, being an acid solution, will rather dye wool and polyamide. Color strength for wool reaches values of 21.01 when the volumetric ration is 9 mL mordant: 3mL crude extract and 27.62 when one only uses 5 mL mordant and 14 mL crude extract. As the dyeing liquor *p*H becomes acid (when the volumetric ratio mordant: crude extract = 5:14), the color strength of the dyed samples increases.

The same effect is also obtained for polyamide, but the K/S values vary between 8.56 and 11.84. The increase of the volume of crude extract used as dyeing liquor leads to the increase of color strength. The obtained values of K/S do not exceed 1.61 on cotton and 0.76 on cellulose acetate.

FT-IR

Lanski and Newman [30] indicated that the pomegranate peel from Punica granatum has a complex structure, including the following components: Hydroxybenzoic acids (*Gallic acid, Ellagic acid*), Hydroxycinnamic acids (*phenylpropanoids: Caffeic acid, Chlorogenic acid, p-Coumaric acid*), Flavan-3-ols (*Flavan-3-ol, Catechin, Epicatechin, Epigallocatechin 3-gallate*), Flavonols (*Quercetin, Kaempferol*), Flavones (*Luteolin, Luteolin 7-O-glycoside*), Flavanone glycoside (*Naringin*), Ellagitannins (*Punicalin, Punicalagin, Corilagin, Casuarinin, Gallagyldilacton, Pedunculagin, Tellimagrandin, Granatin A, Granatin B*).

To these components one must add the anthocyaninic polyphenols and Pelletierine alkaloids (pseudopelletierine and N-methylgranatonine) [31].

Therefore, the brown color of the crude extract used for dyeing is mainly due to the color mix from the pigments extracted from pomegranate peels. These pigments are in fact some anthrocyanidins of delphinidin (blue-red), cyaniding (orange-red) and pelargonidin (orange) type. The N-methyl granatonine is colorless, but it becomes yellow at exposure [31], and the extracted tannins are usually brown.

FT-IR analyses confirm the presence on the textile materials of colored compounds extracted from pomegranate peels, because the absorption bands of the dyed samples are more intense than for the untreated wool (fig. 3) and polyamide (fig. 4).

On the untreated wool FT-IR spectrum (fig. 3) there are the following peaks [32-37]: 3394 cm^{-1} (N-H and O-H stretchings), 2929 cm^{-1} (C-H stretching), 1650 cm^{-1} (amide I), 1511 cm^{-1} (N-H bending from amide II), 1455 cm^{-1} (aromatic in ring, Ar C-C stretch), 1382 cm^{-1} (C-N stretching of amines), 1235 cm^{-1} (C-N stretching from amides III), 1069 cm^{-1} (C-O stretching) and 657 cm^{-1} for combination of N-H wag amines with C-H out of plane from substituted aromatics of some aminoacids in wool.

The FT-IR of untreated polyamide (fig. 4) indicates the presence of the following peaks [38, 39]: 3308 cm^{-1} (N-H free specific band from the polyamide (axial deformation), 2928-2863 cm^{-1} (CH$_2$ antisymetric and symmetric stretchings), 1647 cm^{-1} (C=O stretching, amide I), 1539 cm^{-1} (N-H deformation (from amide II) and C-N stretching vibrations), 1459 cm^{-1} (CH$_2$ deformation), 1369-1118 cm^{-1}

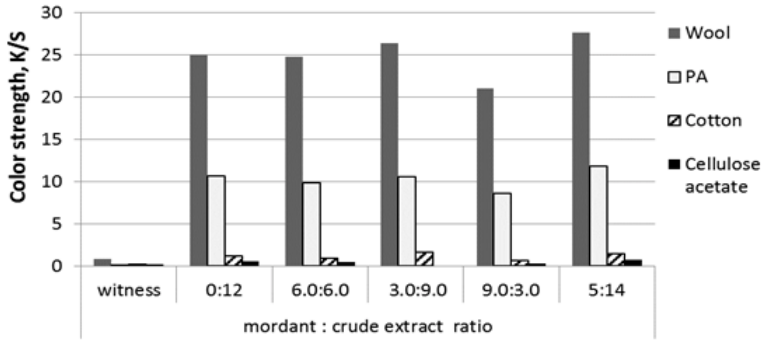


Fig. 2. Color strength for samples dyed with crude extract from pomegranate peel

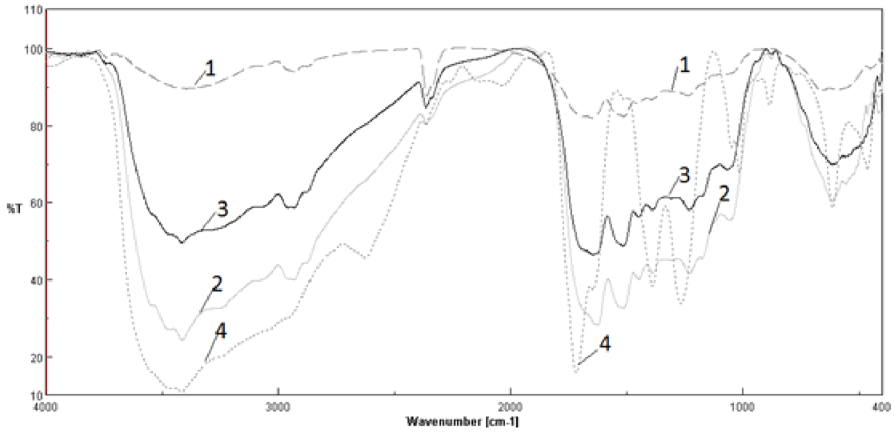


Fig. 3. FT-IR spectra for wool samples dyed with extract from pomegranate peel: (1)untreated wool (1); 0:12 W sample (2); 6:6 W sample (3); crude extract (acid colored solution) (4)

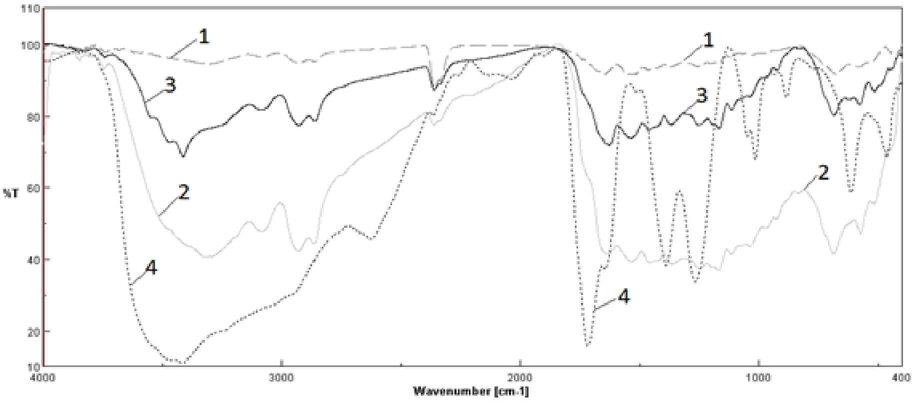


Fig. 4. FT-IR spectra for polyamide samples dyed with extract from pomegranate peel: untreated polyamide (1); 0:12PA sample (2); 6:6 PA sample (3); crude extract / acid colored solution obtained at the end of pomegranate peel maceration by means of the acetic acid (4)

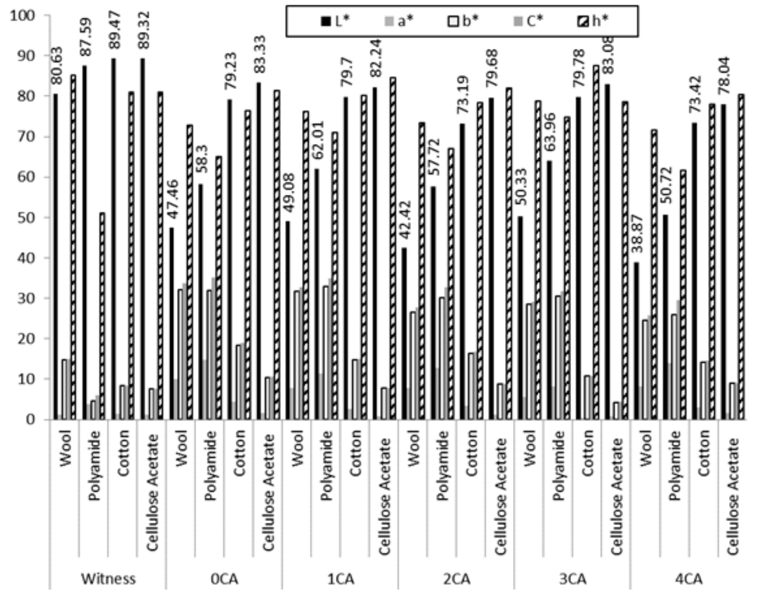


Fig. 5. CIE L*a*b* measurements for the samples dyed with brown extract obtained from pomegranate peels maceration by means of glacial acetic acid

(amide III band involves contributions from both the C-N stretching and N-H bending vibrations) and 678-582 cm^{-1} (N-H wag amines).

The IR analysis confirms the presence of some colored components on the surface of dyed wool/polyamide samples, even if the spectra preserve the characteristic aspect of the witness samples.

The pigments presence on the dyed samples is confirmed by the presence of some absorption bands more intense within the range 3600-3000 cm^{-1} (figs. 3 and 4). responsible for these modifications. Responsible for these modifications are the groups OH, Ar ring, Ar-OH, CH_3= O, COOH (from anthrocyanidins or tannins) and CH_3, piperidine rings, -NH groups from the structure of N-methyl granatonine.

One can notice from figures 3 and 4 that the spectra of wool and polyamide samples dyed with crude extract of pomegranate peel (0:12W and 0:12PA) without mordant in the liquor bath led to the most intense absorption bands. One can explain this through the big volume of the crude extract used for dyeing (12 mL). Mordant utilization (6 mL) only determined the decrease of the peaks size/height, preserving the spectrum aspect, as in the previous case. This can be the result of the utilization of a smaller volume of dyes/crude extract (yet equal to that of mordant (6 mL)) as well as to the effect of dyes fixation on the textile materials, effect generated by the utilized mordant.

*CIE L*a*b* measurements*

The colorimetric measurements indicated that all the dyed samples have smaller lightness than the afferent witness samples. It is known that a small lightness value (as compared to the witness) is equivalent to the intense dyeing of the corresponding sample. In figure 3, one can notice that at lightness hierarchic order is the same for wool and polyamide samples, namely: witness >3CA>1CA>0CA>2CA>4CA. Cotton and cellulose acetate samples are dyed very little, therefore their lightness values are close to those of the afferent witness. It is a fact that cotton can be dyed with direct dyes in neutral medium; or with reactive/vat dye in alkaline medium; yet, the colored solution extracted from pomegranate peel is strongly acid due to the presence of the solvent/glacial acetic acid.

Neither the cellulose acetate samples were intensely dyed, even if the acid medium was ensured, being well known that this material dyes usually with dispersion dyes in acid medium. One can infer that the natural extracted dyes do not belong to the class of the dispersion dyes. They behave similar to acid dyes, because they dye very well both the wool and the polyamide.

What concerns the hues of the obtained colors, one can notice that all the dyed samples are redder (they have a* > 0) than the afferent witnesses, but also more yellow (they have b* > 0).

*CIE L*a*b* color differences*

All the wool samples were intensely dyed by the extract of pomegranate extract (table 2), fact confirmed by the very high values of ΔE* (ΔE* = 33.536 to 43.408 AN units). All the dyed wool samples have ΔL* < 0, which means that they dyed more intensely than the witness. As Δa* > 0 and Δb* > 0, it follows that the brown dyed wool has red reflexes/shades, as well as marked yellow reflexes/shades, which confirms the presence of pigments (anthrocyanins and alkaloid form) in the dyed materials.

Table 2

COLOR DIFFERENCES OBTAINED BY COMPARING DYED WOOL COLOR WITH WITNESS COLOR

M : CE^a (mL:mL)	Wool codes	ΔE*	ΔL*	Δa*	Δb*	ΔC*	Δh*	Observations
0:0	0:0W	-	-	-	-	-	-	
0:12	0:12W	38.403	-33.176	8.659	17.296	18.741	-4.787	darker redder yellow
6:6	6:6W	36.346	-31.549	6.464	16.850	17.726	-3.390	
3:9	3:9W	40.506	-38.209	6.606	11.709	12.799	-4.116	
9:3	9:3W	33.536	-30.308	4.411	13.662	14.168	-2.315	
5:14	5:14W	43.408	-41.768	6.839	9.642	10.896	-4.585	

^{a}M : CE is the volumetric ratio for mordant: crude extract

Table 3

COLOR DIFFERENCES SPECIFIC TO DYED POLYAMIDE SAMPLES

M : CE^a (mL:mL)	PA codes	ΔE*	ΔL*	Δa*	Δb*	ΔC*	Δh*	observation
0:0	0:0PA	-	-	-	-	-	-	
0:12	0:12PA	41.518	-29.296	11.097	27.246	29.206	3.533	darker redder yellow
6:6	6:6PA	38.904	-25.583	7.527	28.327	28.875	5.033	
3:9	3:9PA	40.288	-29.868	8.968	25.507	26.753	3.915	
9:3	9:3PA	35.394	-23.636	4.523	25.953	25.724	5.686	
5:14	5:14PA	43.765	-36.876	10.255	21.223	23.445	2.431	

^{a}M : CE is the volumetric ratio for mordant: crude extract

Table 4

COLOR DIFFERENCES SPECIFIC FOR DYED COTTON SAMPLES

M : CE^a (mL:mL)	Cotton codes	ΔE*	ΔL*	Δa*	Δb*	ΔC*	Δh*	Observation
0:0	0:0C	-	-	-	-	-	-	
0:12	0:12C	14.719	-10.240	3.122	10.101	10.525	-1.006	darker redder yellow
6:6	6:6C	11.718	-9.773	1.205	6.353	6.465	-0.149	
3:9	3:9C	18.271	-16.277	2.024	8.050	8.283	-0.527	
9:3	9:3C	10.054	-9.694	-0.853	2.527	2.433	1.092	darker less red yellow
5:14	5:14C	17.166	-16.046	1.703	5.857	6.071	-0.585	darker redder yellow

^{a}M : CE is the volumetric ratio for mordant: crude extract

Table 5

COLOR DIFFERENCES SPECIFIC FOR CELLULOSE MONO-ACETATE DYED SAMPLES

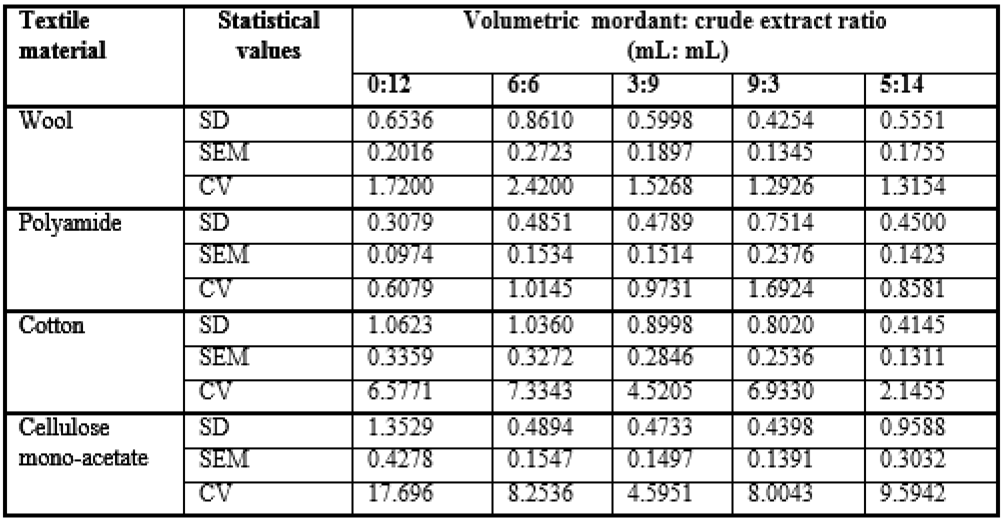
M : CE^a (mL:mL)	cellulose mono-acetate codes	ΔE*	ΔL*	Δa*	Δb*	ΔC*	Δh*	Observation
0:0	0:0CMA	-	-	-	-	-	-	-
0:12	0:12CMA	6.656	-5.984	0.392	2.887	2.913	0.053	darker redder yellow
6:6	6:6 CMA	7.102	-7.080	-0.450	0.320	0.262	0.486	darker less red yellow
3:9	3:9 CMA	9.709	-9.635	0.044	1.198	1.191	0.133	darker yellow
9:3	9:3 CMA	7.065	-6.242	-0.330	-3.292	-3.300	-0.248	darker less red less yellow
5:14	5:14 CMA	11.36	-11.273	0.342	1.433	1.469	-0.105	darker redder yellow

^{a}M : CE is the volumetric ratio for mordant: crude extract

Polyamide behaves similar to wool during dyeing, due to their similar chemical structure (they have polypeptidic/amidic chain and COOH and NH_2 groups as final functional groups). Therefore, the remarks made at wool samples are also valid in the case of dyed polyamide samples; yet, even if they dye more intensely than wool (they have $\Delta E^*_{polyamide} > \Delta E^*_{wool}$, the last ones seem to be lighter, due to more intense yellow hues ($\Delta b^*_{polyamide} > \Delta b^*_{wool}$). The hues difference can be also noticed from the values of Δh* parameter. All the dyed wool samples have $\Delta h^* < 0$, while all the polyamide samples have $\Delta h^* > 0$.

Cotton and cellulose acetate dyed less than wool and polyamide. Perhaps the colored compounds/pigments extracted from pomegranate peels are not specific/indicated for dyeing the cellulose materials or those of esterified derivates. Tables 4 and 5 present the color differences noticed by comparing the dyed samples with witness samples. The last column from tables 4 and 5 specify the conclusions on the dyeing capacity of each sample (*darker*) and hues (from r*edder yellow* to less red *yellow*).

Table 6
STATISTIC ANALYSIS OF THE TOTAL COLOR DIFFERENCE, ΔE*



Textile material	Statistical values	Volumetric mordant: crude extract ratio (mL: mL)				
		0:12	6:6	3:9	9:3	5:14
Wool	SD	0.6536	0.8610	0.5998	0.4254	0.5551
	SEM	0.2016	0.2723	0.1897	0.1345	0.1755
	CV	1.7200	2.4200	1.5268	1.2926	1.3154
Polyamide	SD	0.3079	0.4851	0.4789	0.7514	0.4500
	SEM	0.0974	0.1534	0.1514	0.2376	0.1423
	CV	0.6079	1.0145	0.9731	1.6924	0.8581
Cotton	SD	1.0623	1.0360	0.8998	0.8020	0.4145
	SEM	0.3359	0.3272	0.2846	0.2536	0.1311
	CV	6.5771	7.3343	4.5205	6.9330	2.1455
Cellulose mono-acetate	SD	1.3529	0.4894	0.4733	0.4398	0.9588
	SEM	0.4278	0.1547	0.1497	0.1391	0.3032
	CV	17.696	8.2536	4.5951	8.0043	9.5942

Statistic analysis of the dyeing capacity

The capacity of textile materials to get dyed with brown extract obtained during pomegranate peels maceration with glacial acetic acid was analyzed statistically by calculating SD, SEM and CV for ΔE* parameter (table 6).

Data from table 6 indicate small values for SD, SEM and CV at wool and polyamide samples, which means that the samples from each set of 10 samples from each treatment behaved similarly during the dyeing, acquiring very close colorimetric parameters; it follows, indirectly, that dyeing uniformity is good.

For cotton and cellulose acetate samples were obtained higher values for SD, SEM and CV, because these materials are more difficult to dye with the pigments extracted from pomegranate peels; they function similar to the dyes with poor affinities for cotton/ cellulose acetate.

Conclusions

The crude brown extract obtained from pomegranate peels maceration, in the presence of glacial acetic acid, dyes very intensely wool and polyamide, and less the cotton and cellulose acetate. Utilization of the brown extract without resorting to a solvent/acetic acid separation favors the dyeing of wool/polyamide, because this needs an acid medium for dyeing; perhaps the extracted dye compounds function as acid dyes, this being the only dye class that dye both wool and polyamide. The presence of colored compounds/pigments on the dyed samples is confirmed by the spectroscopic analysis in the IR range. Spectra of all the dyed samples indicate amplifications of the absorption bands specific for the extracted pigments (anthrocyaninic and alkaloid types). Colorimetric measurements (color strength, CIE L*,a*,b* color and CIE L*a*b* color differences) also indicate a high dyeing capacity of wool and polyamide, capacity that increase as the brown extract volume/colored solution increases. The statistic analysis of ΔE* values indicates small color differences on each set of 10 tested samples, for dyed wool and polyamide samples. This fact also indicates a good dyeing uniformity. In exchange, for the cotton and cellulose acetate samples, the statistic analysis indicate higher values for SD, SEM and CV, which indicates a poorer dyeing capacity, generated by poorer affinities of the natural pigments for these materials, and a much poorer dyeing uniformity.

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