Cotton screen-printing with flavonoid natural dyes – Part I. Extraction and application

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In view of the development of awareness of environmental protection and circular economy and the increasing interest in natural dyes, the aim of this work was to investigate the process parameters for screen printing of cotton with natural dyes extracted from onion skin and pomegranate peel. The viscosity of Prisulon DCA 90, a universal thickener for textile printing, was tested under acid, neutral and alkaline conditions. Printing was carried out with acid, neutral and alkaline printing pastes with and without metal salts. The results were evaluated spectrophotometrically. It was found that screen-printing with natural dyes of plant origin can be used for cotton fabrics, achieving a wide range of colour hues under different pH conditions, and the addition of metal salts what significantly improves the colour fastness too.

Key words: *natural dyes; flavonoids; screen-printing; thickener; viscosity; metal salts.*

Izvorni znanstveni rad

Obzirom na razvoj svijesti o zaštiti okoliša i kružnom gospodarstvu te sve većeg interesa za prirodna bojila, cilj ovog rada bio je istražiti procesne parametre za sito-tisak pamuka prirodnim bojilima ekstrahiranim iz ljuski luka i kore nara. Viskoznost Prisulona DCA 90, univerzalnog ugušćivača za tisak tekstila, ispitana je u kiselim, neutralnim i alkalnim uvjetima. Tisak je proveden kiselim, neutralnim i alkalnim pastama s i bez metalnih soli. Rezultati su vrednovani spektrofotometrijski. Utvrđeno je da se sito-tisak s prirodnim bojilima biljnog podrijetla može koristiti za pamučne tkanine, postižući široku paletu nijansi boja u različitim pH uvjetima te dodatkom metalnih soli što značajno poboljšava i postojanost obojenja.

Ključne riječi: prirodna bojila; flavonoidi; sito-tisak; ugušćivač; viskoznost; metalne soli.

1. Introduction

Most natural textile dyes of plant origin belong to the flavonoid group. Flavonoids are a group of polyphenolic compounds found in many plants, especially in seeds, fruit peel, tree bark, leaves and flowers. They were discovered in 1930 by Nobel Prize winner Albert von Szent-Gyorgyi Nagyrapolt, and to date more than 6,400 flavonoids have been identified [1-3]. The multifunctional properties of textiles treated with plant extracts are based on the properties of flavonoids. Many therapeutic effects are attributed to flavonoids, for example antibacterial, anti-inflammatory, anti-allergic, antimutagenic and anticarcinogenic [1, 4-8]. These compounds play an important role in maintaining and protecting the vital functions of plants, and through food they play a similar role for other living organisms. In addition, they are also responsible for protection against UV radiation [9-11].

The use of flavonoids to achieve coloured effects on textiles is based on their chemical structure. The basic structure of flavonoids consists of diphenylpropane, i.e. 1-phenyl-3-(2-hydroxyphenyl)propan-1-ol, from which the basic structure of flavan (Fig.1) is formed by extracting a water molecule and closing the ring, from which other structures of flavonoids are derived.

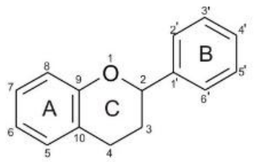


Fig.1 Chemical structure of flavan [1]

These polyphenols consist of two aromatic (A and B) and one heterocyclic ring (C), which are hydroxylated (R-OH) or methoxylated (R-O-CH_3). Relatively 90 % of flavonoids occur in nature in the form of glycosides. Due to the glycosidic bond (R-OH), they are susceptible to cross-linking and polymerisation (e.g. tannin) [1, 2].

Looking at the dyeing properties, most natural dyes are classified as mordant dyes, some as vat dyes and, more rarely, in the group of substantive and basic dyes [2, 3]. Mordant dyes produce different colours by complexing with metal salts (mordants), usually aluminium, copper and iron salts. In view of the chemical properties of flavonoid derivatives, natural dyes of plant origin have historically been used for dyeing protein textile materials (wool, silk) [2, 3, 12, 13]. However, the research presented in this paper represents an important contribution to the demands of contemporary fashion design and market trends that emphasise the use of natural dyes for cellulosic materials. In addition, research has progressed in the field of textile printing of cotton material based on the properties of the chemical compounds of flavonoids. For this reason, plants rich in the flavonoid derivatives quercetin (Fig.2) from onion skin [4, 6, 14, 15] and punicagalin (Fig.3) and punicalin (Fig.4) from pomegranate peel [2, 5, 14, 17-19] have been studied.

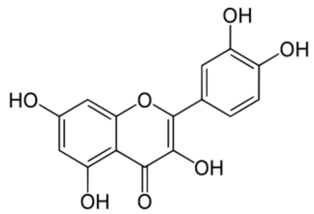


Fig.2 Quercetin [2]

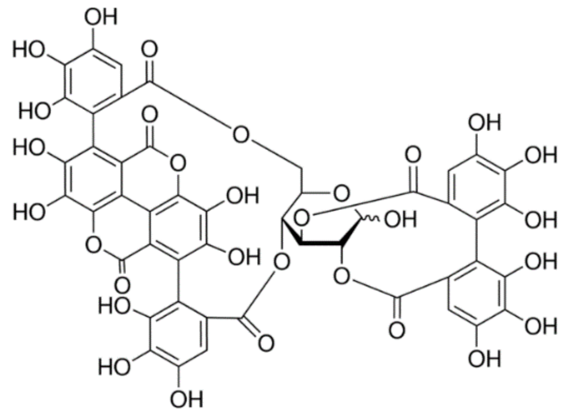


Fig.3 Punicagalin [2]

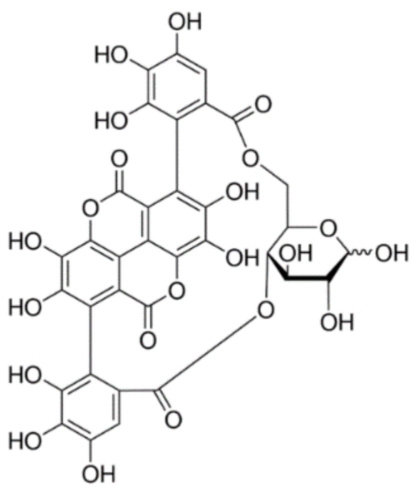


Fig.4 Punicalin [2]

The colouration of the cotton materials and their properties are the result of the formation of ligands between the hydroxyl group of the cellulose, metal ions and hydroxyl or carbonyl groups of the flavonoid compounds (Fig.5) [13, 20, 21].

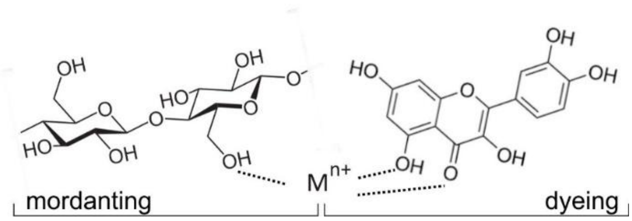


Fig.5 Schematic representation of the formation of the ligand: cotton fiber – metal ion – natural dye [13]

Considering that glycosides are easily hydrolysed at high temperatures under acid and alkaline conditions and under the action of enzymes [1], pH conditions are an important parameter in the application of these compounds for dyeing and printing textiles, both from the point of view of achieving a wide tonality and improving fastens properties [22-25].

2. Experimental

2.1. Textile material

It was used 100 % chemically bleached cotton fabric in plane weave from producer Čateks d.o.o., Čakovec, Croatia. Textile has the following structural characteristics: weight 191.45 g/m^2, density in warp direction 26 cm^{-1}, density in weft direction 25 cm^{-1}.

2.2. Dyestuff extraction

The textile material was printed with plant extracts from onion skin and pomegranate peel collected in Croatia. Dye extraction was performed in soft water (1.27 µS) containing 20 g/L of each plant. The extraction was performed in a 1:40 bath ratio (considering the mass of the plant) at 100 °C for 60 min. The bath was then allowed to cool for 12 h and the extract was decanted. In addition, the extract obtained was decanted and the water evaporated to obtain a dry natural dye.

2.3. Thickener analyses

Prisulon DCA 90 is guar ether universal thickener for textile printing on all kinds of fibres and with all dyestuff classes. Well stable to water hardness, bivalent and trivalent metal salts and all common printing auxiliaries [26]. Thickener was examined with dry substance content of 9 %, and was tested at three different pH values: acidic, pH 5 adjusted with 20 % acetic acid (Kemika, Zagreb, Croatia), neutral, pH 7 and alkaline, pH 10, adjusted with Na_2CO_3 (Kemika, Zagreb, Croatia). The viscosity of the thickener was determined using a Brookfield DV II+ viscometer, LV4 spindle. The measurement was carried out at a constant temperature 20±0.3 °C.

2.4. Printing paste preparation

Printing paste recipes was as follow: 100 g thickener, 2 g natural dye, 0.5 g urea, 0.2 g glycerol, 0.1 g metal salt, and pH regulator. For each plant were prepared acidic, neutral and alkaline printing pastes without metal, and with metal salts: potassium aluminium sulphate dodecahydrate $KAl(SO_4)_2 \cdot 12H_2O$, copper(II) sulphate pentahydrate $CuSO_4 \cdot 5H_2O$, and ferrous(II) sulphate heptahydrate $FeSO_4 \cdot 7H_2O$ (Kemika, Zagreb, Croatia).

2.5. Screen-printing process

The printing process was conducted by manual screen-printing, using the following tools: a manual lab-type flat screen-printing table, a wooden-handled polyurethane blade squeegee of 60 durometer hardness, and an aluminum 40 × 50 cm screen with a mesh of 62 threads/cm^2. Considering that the applied method was hand screen-printing, the squeegee angle was kept between 7-10°, with optimal pressure to assure uniform transfer of the printing paste to the surface of a textile substrate. Fixing was carried out with a heat press at 120 °C, 5 minutes. After fixing procedures, the samples were rinsed in warm and cold water.

2.6. Colour Analysis

Colour characteristics were determined using a remission spectrophotometer, Datacolor 850, measuring geometry d/8°, illumination D65, and measuring aperture of 9 mm. The coordinates used to determine colour values are L* for lightness, a* for redness (positive value) and greenness (negative value), b* for yellowness (positive value) and blueness (negative value), C* for chroma and h° for hue angle in the range of 0° to 360°. Colour coordinates of undyed and dyed cotton fabrics were determined according to ISO 105-J01:1997 *Textiles — Tests for colour fastness — Part J01: General principles for measurement of surface colour*. All results were measured on samples by repeating the measurement procedure at random locations. Thus, the colour measurements were made using the Datacolor Tools computer program and “Measuring until tolerance” command, which means that at least 10 measurements must be made, and the results are accepted only if the total colour difference between each measurement is less than 0.1 (dE < 0.1).

2.7. Colour fastness to laundering

Colour fastness to laundering of printed materials were tested in a laboratory apparatus for wet processes Polycolor (Mathis, Switzerland). The test was performed according to standard ISO 105-C06:2010 (A2S) *Textiles — Tests for colour fastness — Part C06: Colour fastness to domestic and commercial laundering*, using 2 g/l of standard detergent (James Heal ECE A, without optical brighteners and without phosphates), with a bath ratio of 1:20, temperature of 40 °C, time of 30 min. The results of fastness to laundering are given as numerical values of total colour difference (dE) calculated according to equation (1):

$$dE = ((dL^*)^2 + (da^*)^2 + (db^*)^2)^{1/2} \qquad (1)$$

where:
dL^* = (L^* treated) – (L^* untreated);
da^* = (a^* treated) – (a^* untreated);
db^* = (b^* treated) – (b^* untreated).

2.8. Colour fastness to artificial light

Colour fastness to artificial light were performed on Xenotest 440 ((SDL Atlas, Rock Hill, SC, USA). Xenotest 440 is used for laboratory simulation of external weather influences on the stability and durability of textile and other materials. Analysis was evaluated according to the modified ISO 105-B02 and 13 B04 test methods using Xenotest 440. Test conditions simulated in this research were: Total light time: 41:10 h, Radiant exposure: 6226 kJ/m^2, Irradiance control: 300–400 nm, Filter system: B04, E: 42 W/m^2 (± 2 W/m^2), CHT: 32°C (± 3°C), BST: 47 °C (± 8°C), RH: 40 % (± 8 %), no spray, fan speed: 2000 rpm.

Using the same equation (1) as for the fastness to laundering, the artificial light fastness properties was also evaluated by calculating total colour difference values (dE).

3. Results and discussion

In order to test the pH value of the printing paste as a parameter, it was important to determine the stability of the thickener under different pH conditions. An analysis of the viscosity properties of the thickener Prisulon DCA 90 thickener is shown in Tab.1.

The viscosity measurements carried out for the Prisulon DCA 90 thickener at a solids content of 9 % show a slight difference in viscosity depending on the pH change at the same spindle speed, i.e. at the torque values. Deviations in viscosity values are satisfactory realised for manual screen printing. Under the given conditions, the highest applicable spindle speed was 12 rpm, with a viscosity of 283.9 mPas in the acid pH, 216.5 mPas in the neutral and 292.9 mPas in the alkaline pH.

Based on the results of measuring the viscosity of the thickener DCA 90, the same thickener was used for the preparation of printing pastes. Flavonoid natural dyes extracted from onion skin and pomegranate peel were used without the addition of metal salts and with the addition of metal salts (Al, Cu, Fe), acidic, neutral and alkaline printing pastes were prepared in all combinations.

The visual evaluation of the printed cotton samples is shown in tab.2 and 3. The visual assessment of the sample is extremely important for the use of natural dyes, as it often deviates considerably from objective spectrophotometric measurements. It is important in the application of natural dyes, especially in the fashion industry or in textile restoration. It should be noted that by changing both parameters, the pH conditions and the emphasised use of metal salts, a wide range of yellow hues can be achieved.

Tab.1 Viscosity of the thickener Prisulon DCA 90

RPM	pH 5			pH 7			pH 10		
	h (mPas)	t (%)	T (°C)	h (mPas)	t (%)	T (°C)	h (mPas)	t (%)	T (°C)
0.3	439.9	2.1	20.3	379.9	1.9	20.1	419.9	2.1	20.1
0.6	539.9	5.5	20.2	459.9	4.6	20.0	579.9	5.7	20.2
1.5	515.9	12.9	20.0	387.9	9.7	20.0	515.9	12.9	20.2
3	433.9	21.6	20.2	323.9	16.3	20.1	441.9	22.1	20.2
6	355.9	35.7	20.1	267.9	26.9	20.1	365.9	36.6	20.3
12	283.9	56.8	20.3	216.5	43.4	20.1	292.9	58.9	20.3
30	E	E	E	160.9	80.0	20.1	E	E	E
60	E	E	E	E	E	E	E	E	E

E – outside the range of the measuring instrument

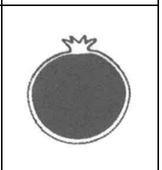
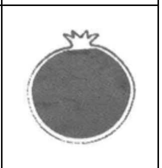
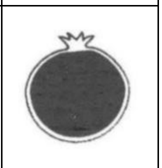
Without the metal ions and with aluminum, brighter colour tones are obtained, with copper duller tones and with the addition of iron very dark patterns. The samples shown in tabs 2 and 3 also confirm the good selection of the universal guar ether thickener with a dry substance content of 9 % and the fixation with a heat press at 120 °C for 5 minutes, which ensures brilliant colouration, good surface coverage and sharp contours.

The spectrophotometric analysis of cotton samples printed with natural dyes extracted from onion skin and pomegranate peel, with acidic, neutral and alkaline paste without and with addition of metal salts is shown in tabs 4 and 5. Considering the content of flavonoids in the selected plants, the expected colour hues in the yellow-orange range was obtained.

Tab.2 Cotton-onion samples printed with acid, neutral and alkaline paste

pH	Metal ions			
	-	Al	Cu	Fe
5				
7				
10				

Tab.3 Cotton-pomegranate samples printed with acid, neutral and alkaline paste

pH	Metal ions			
	-	Al	Cu	Fe
5				
7				
10				

Tab.4 Colour parameters of cotton-onion samples printed with acid, neutral and alkaline paste

Metal ions	L*	a*	b*	C*	h°
pH 5					
-	71.03	18.38	24.33	30.49	52.93
Al	74.50	11.39	36.42	38.16	72.63
Cu	66.05	11.38	28.04	30.26	67.92
Fe	48.27	2.21	10.59	10.82	78.23
pH 7					
-	73.04	17.71	23.66	29.56	53.18
Al	70.99	13.34	38.45	40.70	70.86
Cu	66.66	10.31	25.74	27.73	68.18
Fe	50.83	2.03	9.89	10.09	78.39
pH 10					
-	70.03	18.38	24.33	35.19	50.13
Al	69.14	16.85	36.26	39.98	65.08
Cu	63.42	9.03	23.98	25.62	69.36
Fe	42.66	3.00	9.66	10.12	72.75

Tab.5 Colour parameters of cotton-pomegranate samples printed with acid, neutral and alkaline paste

Metal ions	L*	a*	b*	C*	h°
pH 5					
-	75.65	5.01	30.88	31.29	80.79
Al	75.93	3.69	36.29	36.48	84.20
Cu	63.06	6.23	26.02	26.75	76.54
Fe	45.95	-0.60	4.31	4.35	97.90
pH 7					
-	75.47	5.30	30.62	31.08	80.19
Al	75.22	4.15	37.94	39.16	83.92
Cu	63.75	5.81	25.67	26.32	77.25
Fe	51.30	-0.50	6.16	6.18	94.68
pH 10					
-	70.09	6.90	32.82	33.54	78.12
Al	71.33	5.34	39.66	40.02	82.34
Cu	64.29	5.36	24.24	24.73	77.48
Fe	40.57	0.38	6.11	6.12	86.47

Quercitin (Fig.2) from onions gives an orange colour hue around 50°, while the addition of aluminium shifts this value towards 70°. The addition of copper has a greater effect on the change in lightness, so yellow-brownish colouration is obtained. Iron affects both the reduction in lightness and the colour chromaticity value, which decrease from 30 to just 10, resulting in dark, brown-black samples.

The flavonoids punicagalin (Fig.3) and punicalin (Fig.4) from pomegranate produce yellow colour hues around 80°. Their reaction with aluminium shifts the h° value slightly towards the yellow range. However, aluminium increases the colour hue value in all samples, and the use of this salt results in a more brilliant colour. The reaction of these flavonoids with cupper ions leads to the formation of a colour complex with a lower colour lightness and the colouring is more subdued. A colour hue above 85° when using iron salt does not come to the fore, as all samples appear black due to the low chromaticity, which is below 7. The changes in the colour parameters are more pronounced with acidic and alkaline printing paste, as according to Grotewold [1] a hydrolysis of the glycosides takes place, i.e. their cross-linking decreases and they become more reactive.

The colour fastness of the samples to laundering (Fig.6 and 7) and artificial light (Fig.8 and 9) was calculated according to Eq (1) as the value of the total colour difference (dE) of the treated samples compared to the untreated samples.

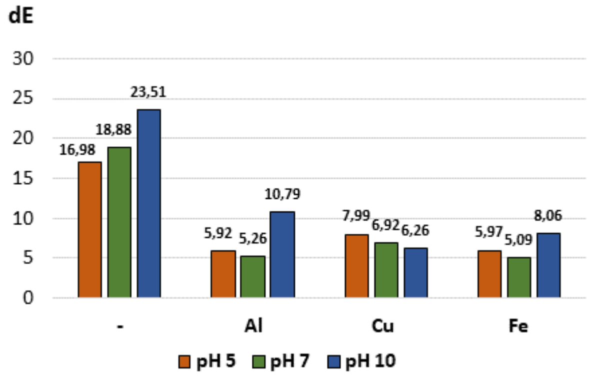


Fig.6 Fastness to laundering of cotton-onion samples printed with acid, neutral and alkaline paste

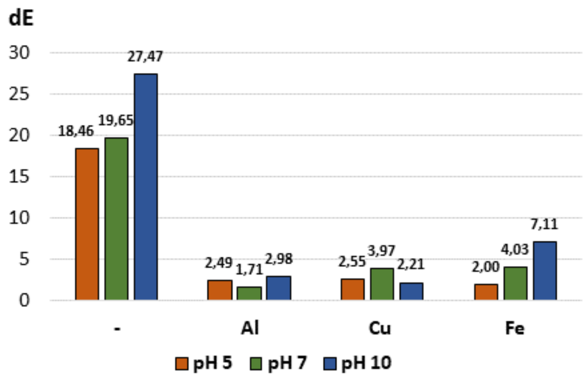


Fig.7 Fastness to laundering of cotton-pomegranate samples printed with acid, neutral and alkaline paste

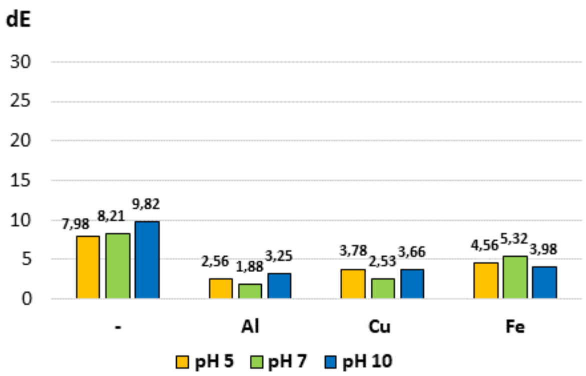


Fig.8 Fastness to artificial light of cotton-onion samples printed with acid, neutral and alkaline paste

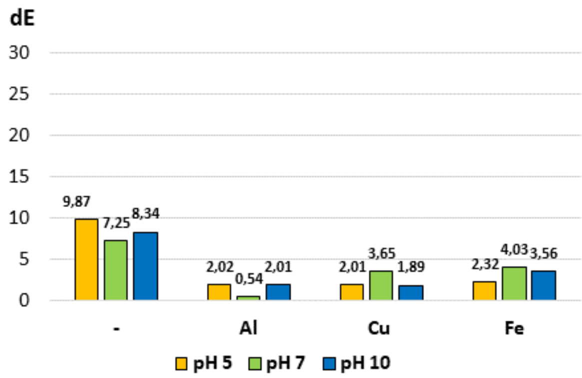


Fig.9 Fastness to artificial light of cotton-pomegranate samples printed with acid, neutral and alkaline paste

The graphs clearly show that the addition of metal ions to the printing paste and the formation of the ligand: cotton fiber – metal ion – natural dye (Fig.5) plays a greater role for the colour fastness than the pH values. Samples printed without metal have the lowest fastness, i.e. the highest total color difference (dE), more than 16 for fastness to laundering, and above 7 for fastness to light. However, in general, it can be said that the printed causes obtained with alkaline printing paste can have worse performance properties, it can be observed that the samples have a weak fastness on laundering and the dE values are above 15. When using metal in acidic and neutral printing paste, they fall below 8 for the dye extracted from onions (Fig.6) and below 4 for pomegranate (Fig.7). The light fastness is better and dE is below 10 and with the addition of metal below 5 (Fig.8 and 9).

4. Conclusion

The investigation of flavonoid natural dyes for use on cotton material in the screen-printing technique confirmed that they can be successfully used with the universal thickening agent Prisulon DCA 90 guar ether with a dry substance content of 9 % and the fixation with a heat press at 120 °C for 5 minutes. The viscosity of the selected thickener is satisfactory for this technique under acidic, neutral and alkaline conditions. By varying the pH values of printing pastes, the influence on the molecules of the flavodnoid derivatives of quercetin extracted from onion skin and punicagalin and punicalin extracted from pomegranate peel was confirmed. This influence is most pronounced under alkaline conditions and in terms of lightness and chromaticity values as well as weaker fastness to laundering and artificial light. The greatest influence on the results is the influence of the metal salts of aluminium, copper and iron on the colour parameters and on the satisfactory achievement of laundering and light fastness. This study has confirmed that natural flavonoid dyes can be successfully used for screen-printing of cotton fabrics for obtaining brilliant colours, sharp contours and good colour fastness.

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