ANTIBACTERIAL AND UV PROTECTIVE EFFECTS OF COTTON FABRICS DYED WITH RESEDA LUTEOLA EXTRACT

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Reseda luteola (Weld) extracts were used to dye textiles and decorate medieval manuscripts. The main components are luteolin and apigenin in the form of β-glucosides, having an important role in plant UV protection, coloration and defense. Luteolin and apigenin have anti-oxidant, anti-carcinogenic, anti-inflammatory and anti-bacterial properties. The present study investigates the degree of exhaust of dyestuff extract of Weld in the dyeing process of cotton fabrics pre-mordanted with mimosa tannin and tannin/alum, the antibacterial and UV protective effects of dyed fabrics. The UV-Vis spectra of the dye solution demonstrated the presence of luteolin-7, 3'-di-O'-glucoside as the main compound. The highest dye exhaustion in the dye bath is attained for the cotton pre-mordanted with 8% Mimosa and 15% Alum, probably due to the complex formed between the flavonoids components and Al^{3+} ions. While the ultraviolet protection factor (UPF) for the untreated cotton knit is 10, indicating the lack of protection against UV radiation, all the mordanted and dyed fabrics show an excellent protection (UPF >50+) due to the UV high absorbance of tannins and Reseda luteola components. The only samples demonstrating a satisfactory antibacterial effect against *S. Aureus* are the fabrics pre-mordanted with mimosa tannin and alum and dyed with Reseda luteola extract.

Keywords: dyer's weld, UPF, exhaustion degree.

INTRODUCTION

Reseda luteola L., known as dyer's weld, was used to produce a yellow mordant dye, especially for dyeing wool and silk, as weld lake and for decorating medieval manuscripts (Angelini *et al*., 2003). The main components responsible for the yellow color are luteolin and apigenin. Other components are luteolin glycosides (luteolin 3',7-O-diglucoside, luteolin-7-glucoside, luteolin 4'-O-glucoside), apigenin 7-O-glucoside, kaempferol, quercetin, chrysoeriol (Gaspar *et al*., 2009). Most of the flavonoids are in the form of their β-glucosides, and have an important role in plant UV protection, flower coloration and defense (Schmidt *et al*., 2011). Luteolin and apigenin have anti-allergic, anti-oxidant, anti-carcinogenic, anti-inflammatory and anti-bacterial (Funakoshi-Tago *et al*., 2011; Shukla and Gupta, 2010). The Weld dyestuff (flavonoids) being a mordant dye needs mordants to be fixed on the fiber. The color depends on the metal salts: a lemon yellow is obtained with alum, greenish yellow with copper and olive with iron, deep yellow with slightly hard water or adding sodium carbonate or calcium carbonate to the dyeing bath (Vankar and Shukla, 2018). The present study was carried out to investigate the degree of exhaust of dyestuff extract of Weld (Reseda Luteola) in dyeing process of cotton fabrics pre-mordanted with mimosa tannin and mimosa tannin/alum, the antibacterial and UV protective effects of dyed fabrics.

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EXPERIMENTAL

Materials

Reseda Luteola: Wau-Color - Dyestuff extract of Weld (Reseda Luteola), Dye content: 236g/Kg, kindly provided by NIG Nahrungs- Ingenieurtechnik GmbH, Austria;

Cotton knit, 165 g/m^2, 0.79mm thick, chemically bleached.

Methods

Pre-mordanting: the bleached cotton fabric was pre-mordanted with 2% and 8% mimosa tannin, 2% mimosa and 4% alum, 8% mimosa and 15% alum at 30-40°C for 60 minutes in a liquor ratio of 1:60. After that, the fabrics were maintained at room temperature for 3 hours, washed 3 times with hot, warm and cold water, squeezed and freely dried.

Dyeing: the pre-mordanted fabric was dyed with Reseda Luteola solution (4% o.w.f) in a liquor ratio of 1:60, at 80°C for 1 hour. The squeezed fabric was immersed in 20g/L NaCl and maintained at 40°C for 30 minutes. Then, the fabric was rinsed 3 times with hot, warm and cold tap water, squeezed and dried at room temperature.

The appearance of dyed fabrics is displayed in the table 1.

Table 1. The appearance of the fabrics mordanted with mimosa/alum and dyed with Reseda Luteola

 2% Mimosa	 8% Mimosa	 2% Mimosa 4% alum	 8% Mimosa 15% alum
 Reseda Luteola 2% Mimosa	 Reseda Luteola 8% Mimosa	 Reseda Luteola 2% Mimosa 4% alum	 Reseda Luteola 8% Mimosa 15% alum

Characterisation

UV-Vis spectroscopy (Lambda 950, Perkin Elmer, USA) was used to determine the maximum absorption of weld dye and evaluate the degree of exhaustion representing the amount of dyestuff or mordant diffused in the fibers. Ultraviolet Protection Factor (UPF) was measured on UV-Vis spectrophotometer (Cary 50, Varian, Australia) according to standard AS/NZ 4399:1996. Fabrics are rated as providing an "Excellent UV Protection" if UPF value is 40 or greater, "Good UV Protection" if UPF values are between 25 and 39 and as having "Good UV Protection" if UPF value is in the range 15 – 24 (Hatch, 2003). The samples were tested for microbial activity in *Staphylococcus aureus* according to SR EN ISO 20645/2005. The dyeing fastness was evaluated according the ISO standards, respectively: ISO 105 C06 (washing; ISO 105-E04:2013- (perspiration), ISO 105-X12 (rubbing), ISO 105 B02 (light).

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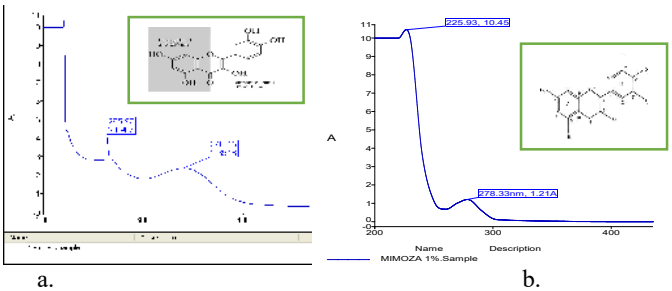
RESULTS

UV-Visible Spectra

The UV-VIS spectrum of Reseda Luteola and mimosa tannin (fig. 1) was recorded to determine the maximum wavelengths absorption.



a. b.

Figure 1. UV-VIS spectrum: a. Reseda Luteola; b. mimosa tannin

The aqueous solution of Reseda Luteola shows 2 absorption bands, situated at 266nm and 340nm, the first being specific for the benzoyl ring and the 2nd for the cinnamoyl structure (Boots *et al*., 2007). By comparisons with literature data, the detected compound belongs to the group of flavonoids and could be luteolin-7,3'-di-O'-glucoside, λmax = 269, 339 nm) (Troalen, 2013) or its mixture with apigenin (λmax = 267, 300, 338 nm). The high solubility in water is another argument for this component knowing that flavonoid glycosylation enhances solubility compared to the corresponding aglycones. 1% Mimosa tannin solution has two maxima absorption, situated at 226nm and 279nm specific for $\pi \rightarrow \pi^*$ transitions in the benzene ring system conjugated with carbonyl group, double bond, or hetero atom (Barnum, 1977).

Degree of Dye Exhaustion

To quantify the dye concentration, a calibration curve (figure 2) was plotted at 340 nm in five points using 0.1g/L, 0.3 g/L, 0.4g/L, 0.5 g/L, 0.7g/L dye solutions.

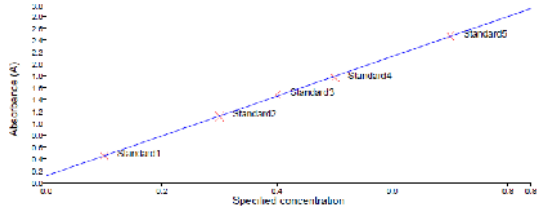


Figure 2. Calibration curve of Dyestuff extract of Weld at 340nm (r^2 =0.9997)

The initial and final concentrations of dye baths were determinate based on calibration curve by recording the absorption of diluted solutions.

The degree of exhaustion was calculated according to equation (1):

$$\% E = 100(C_i - C_f)/C_f \quad (1)$$

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where C_i: the initial concentration; C_f = the final concentration of the dye.

The exhaustion degree of premordanted and dyed cotton fabrics are shown in table 2.

Table 2. The exhaustion degree of the initial and final dye baths, λ =340 nm

Sample	Ci	Cf	Degree of exhaustion, %
4% R. Luteola, 2% Mimosa	0.6273	1.3614	-117.025
4% R. Luteola, 8% Mimosa	0.7158	1.3941	-94.761
4% R. Luteola, 2% Mimosa, 4% Alum	0.993	0.5622	43.383
4% R. Luteola, 8% Mimosa, 15% Alum	1.1136	0.5901	47.009

The negative degree of exhaustion in the case of dyebath used to dye the cotton pre-mordanted with 2% and 8% mimosa tannin demonstrates the low diffusion of dye into fabrics, probably due to saturation of the material with tannin. Additionally, both tannin and flavonoid compounds have similar structures and functional groups, creating an electrostatic repulsion between them. In the case of mordanting with tannin and alum, the exhaustion degree increases, the more the amount of alum is higher. This demonstrates that the agent causing the dye to fix on the material is alum and less, or not at all, tannin. During dyeing, it is possible that the glycosidic compounds are hydrolyzed to parent aglycones (Halpine, 1996). As studies demonstrated both apigenin and luteolin form complex with Al^{3+}, depending on metal ion concentration. Luteolin forms a 2 : 1 complex at low Al^{3+} concentration, a 1 : 1 complex at higher concentration, and a 1 : 2 complex which is in Al^{3+} binds to the 3′,4′-dihydroxyl moiety (Kasprzak *et al*., 2015).

The Colour Fastness Evaluation of the Fabrics Dyed with Reseda Luteola

The colour fastness of the fabrics dyed with Reseda Luteola are shown in table 3.

Tabel 3. Colour fastness of the fabrics mordanted and dyed with Reseda Luteola (RL)

Colour fastness to/Sample	RL + 2% Mimosa	RL + 8% Mimosa	RL + 2% Mimosa + 4% alum	RL + 8% Mimosa + 15% alum
Washing	2	2	1	1
Acid perspiration	2	2	1-2	1-2
Alkaline perspiration	2-3	3	3	3-4
Rubbing dry	4-5	4-5	4-5	4-5
wet	4	4	3-4	3-4
Light fastness	3*	3*	3**	3**

Exposure time: * 26 hours; ** 7 hours

The color change to washing and acid perspiration is very poor, the worst results being recorded for the fabrics mordanted with mimosa and alum. Instead, the fastness to alkaline perspiration is better for the fabrics pre-mordanted with larger amount of mordants, especially with 8% Mimosa + 15% alum. The color change for the dry rubbing is excellent while for wet rubbing is good mainly for the fabric pre-mordanted with 2 and 8% mimosa tannin. The light fastness is moderate.

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Assessment of the Ultra-violet Protection Factor (UPF)

The UPF values of the fabrics pre-mordanted with mimosa tannin and dyed with Reseda Luteola are shown in the table 4.

Table 4. UPF values of the fabrics pre-mordanted and dyed with Reseda Luteola (RL)

Sample	Mean UPF	Mean UVA Transmission	Mean UVB Transmission	UPF calculate	UPF rating
untreated	15.698	11.807	5.289	13.498	10
2% mimosa	136.207	1.345	0.669	114.127	**50+**
8% mimosa	232.204	0.752	0.377	207.556	**50+**
2% mimosa/ 4% alum	187.415	0.849	0.445	157.032	**50+**
8% mimosa/15% alum	189.435	1.084	0.443	154.351	**50+**
2% mimosa /RL	221.794	0.451	0.432	205.066	**50+**
8% mimosa /RL	174.423	0.606	0.559	151.332	**50+**
2% mimosa/ 4% alum / RL	613.328	0.048	0.208	347.282	**50+**
8% mimosa/15% alum / RL	426.001	0.429	0.229	335.383	**50+**

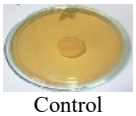
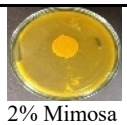
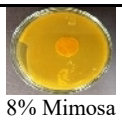
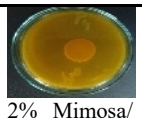
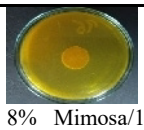
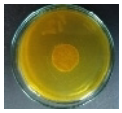
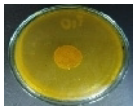
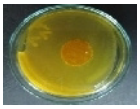
The UPF value for the untreated white cotton knit is 10, indicating the lack of protection against UV radiation. All the treated fabrics provide a protection factor of 50+, ensuring an excellent protection of the human body against harmful UV radiation.

A higher quantity of mimosa tannin lets to pass a smaller amount of UV rays than fabrics mordanted with mimosa/alum. The dyeing further decreases the UVB and transmission UVA through the fabrics, the lowest transmission being ensured by the knit mordanted with 2% mimosa + 4% alum and then dyed with Reseda Luteola.

Antibacterial Activity of Knitted Fabrics Mordanted and Dyed with Reseda Luteola

The cotton fabrics mordanted and dyed were tested against *S. aureus* and the results were compared with control (table 5).

Table 5. Antibacterial effect of mordanted and dyed fabrics

 Control	 2% Mimosa	 8% Mimosa	 2% Mimosa/ 4 % Alum	 8% Mimosa/15% Alum
	 RL/2% Mimosa	 RL /8% Mimosa	 RL/2% Mimosa/ 4% Alum	 RL/8% Mimosa/15% Alum

According to EN ISO 20645:2004 standard, the treatment is considered effective if the inhibition zone is ≥1-0 mm and no growth under specimen is detected, whereas 0 mm inhibition zone and slight growth is evaluated as limited effect. The results show

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the absence of inhibition zone for all the samples. The untreated cotton fabric presents a moderate multiplication of *S. aureus* colonies while the mordanted fabrics and those mordanted with mimosa and dyed show some restricted colonies growth on their surface. The only samples demonstrating a satisfactory antibacterial effect against *S. Aureus* are the fabrics pre-mordanted with 2% Mimosa/ 4% Alum and 8% Mimosa/ 15% Alum and dyed with Reseda Luteola. The improved efficiency could be attributed to the dye if we consider the limited efficacy of the fabrics mordanted with the same mordants.

CONCLUSIONS

The highest dye exhaustion in the dye bath is attained for the cotton pre-mordanted with 8% Mimosa and 15% Alum due to the complex formed between the flavonoids components and Al^{3+} ions. The fabric excellent UV protection is due to the UV high absorbance of tannins and Reseda Luteola components. The low content of dyestuffs on the fabrics decreases the antibacterial efficiency of textiles dyed with Reseda luteola.

Acknowledgments

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