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Eco-friendly printing of natural fabrics via fenugreek gum with safflower, henna and alkanet natural dyes

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Abstract--This work is aimed to use an eco-friendly water soluble gum (extracted from Fenugreek seeds) to investigate its suitability in printing paste as thickening agent with natural dyes named; Safflower, Henna and Alkanet were used as an alternative solution for toxic, carcinogenic and harmful problems resulted from synthetic dyes. The addition of citric acid or sodium carbonate to the printing paste using different values was examined to achieve the best colour strengths on the fabrics and get the most efficiency of the used natural dyes. Alum, Iron, Copper, and Tannic acid were different mordants used to enhance the strength of colours resulted of natural dyes on fabrics.

Keywords--Galactomannan, Citric acid, Sod. Carbonate, Mordant.

1. Introduction

Recently, many countries have issued stricter environmental regulations in response to increase the public concern and awareness. This imposes a lot of pressure on the industry of textile coloration, which is considered one of the most water polluters due to the production of effluent containing dyes and other chemicals. Thus, textile processors have to look carefully at the environmental impact of the products they use. ⁽¹⁷⁾

2- Materials

2.1.1 Fabrics

Pure wool fabric mill scoured produced by Misr Company for Spinning and Weaving; Egypt.

Fabrics specification:

- Weight of square meter = 220 g/m².
- The number of yarn / cm = 24 and 22 for warp and weft respectively.

2.1.2 Pure cotton fabric mill desized, kier boiled and bleached supplied by Misr/Helwan Co. for Spinning and Weaving; Cairo, Egypt.

Fabric specification:

- Weight of square meter = 165 g/m².
- The number of yarn / cm = 40 and 36 for warp and weft respectively.

2.2. Fenugreek seeds (printing paste gum):

Clean, dry fenugreek seeds (*Trigonella foenum-graceum*) cultivate in Egypt, purchased from the local market was used as a raw material in the present work. The gum which extracted from fenugreek seeds is Consists of a galactomannan where galactose and mannose units average ratio of 1:1 attached at the 6-position. Galactomannan is polysaccharide, which is natural, nontoxic and have improved emulsifying properties that appropriate for use in the food and health industries. ⁽⁸⁾ The seeds and leaves, are used to prepare powders or extracts for medicinal use. ^{(1) (15)}

2.3. Natural Dyes

Natural dyes from natural sources which were used in this work were prepared in powdered form from the following plant species.

2.3.1. Safflower:



It is a flowering annual crop, its herb 1 to 1.5 m long. It is also named *Carthamus* may come from hebrew word "kartami" which means to dye, or it is the latinized form of the arabic word *quartum* or *gurtum*, which refers to the pigment colour. ⁽⁷⁾

Safflower was used in dyeing the bandages of Egyptian mummies utilizing the Safflower character to prevent from mold. Also it was and still one of a traditional medicine in China, where it has many utilizations such as immunosuppressant, anticoagulant, antioxidant, vasodilator, and neuroprotector. Moreover, it is widely used in food colorants. ⁽¹³⁾

Some pigments were found in disk floret of Safflower named as: Carthamin, Carthamone, Isocarthamin, Caryophyllene, Caryophyllene-epoxide, Kaempferol - glycoside, and Neocarthamin. Also some minerals were found as: Chromium, Cobalt, Copper, Zinc, Silicon, Phosphorus, Magnesium, Manganese, Iron, Sodium, Potassium, Calcium, Cadmium and Lead.

2.3.2. Henna:



Henna (*Lawsonia inermis*), is a shrub that grow to (2-5) m height with a rough trunk and irregular branching. It can be found in Iran, Pakistan, India and Egypt, but traditionally existed as far East as the Yellow River in China. It is an ancient dye, where the nails of Egyptian mummies which are found in the tombs had dyed with henna. The leaves in aqueous paste or powdered form are used as a cosmetic for palms, hands, hairs. ^{(9) (14)}

The leaves are available commercially whole or in the form of grayish green powder. The Henna flowers are cream, small and extremely have pleasant smelling, similar to honey and were used in cosmetics.

Henna has a dye molecule, Lawsone, also known as hennotannic acid. It is the main colouring ingredient in Henna leaves (2-hydroxy-1-4 naphthoquinone), it is a small molecule and exhibits a remarkable affinity for silk, polyester, nylon and wool. It imparts an orange red colour. And also known to form complexes of different colours with metal salts. It can be applied on wool and silk after pre-treatment to give a range of colours. Lawsone, which has a non-ionic, hydrophobic molecule, very small, show all the characteristics of a disperse dye. ⁽¹⁰⁾ Also the leaves are contain flavonic pigments (luteolin) of yellow colour and tannins which used as organic mordants. ⁽⁹⁾

2.3.3 Alkanet:



Alkanet (*A. tinctoria*) is cultivated in Southern Europe for its dye, which is readily extracted by oils. These herbs comprehended under the name *Anchusa*, to paint or colour anything, Whereupon those plants were called *Anchusa*, that bright and flourishing red colour which is in the root, even as red as pure and clear. This plant gives purples or burgundies in an alkaline dyebath. The color is enhanced by addition of alcohol extract to premordanted wool or silk in dyebath. ⁽¹⁰⁾

All used plant species were purchased from the Egyptian local market.

2.4. Mordants:

Alum, Iron, Copper, and Tannic acid were used as mordants.

2.5. Chemicals:

- Citric acid.
- Sodium carbonate.

- Sodium hydroxide (NaOH).
- Urea.
- Hostapal CV-ET (Hoechst), a nonionic detergent for soaping bath.

All these chemicals were of pure grades.

3- Procedures:

3.1. Preparation of Cotton and wool Fabrics:^{(5) (16)}

The fabrics were washed with 2 g/L Hostapal CV-ET (non-ionic wetting agent) at 45°C for 15 minutes, then were rinsed with warm and cold water respectively to remove any impurities. The prepared fabrics were dried at room temp.

3.2. Extraction of Fenugreek Gum & Printing Paste Preparation:⁽¹⁷⁾

In recent years, galactomannan gum from Fenugreek seeds was used as a printing paste of textile fabrics. One of Fenugreek gum separation methods were selected to separate the endosperm mechanically from the hull and germ which were differ in their hardness properties.

Clean, dry fenugreek seeds were grounded, sieved then soaked overnight. The swelled gum was filtrated through a muslin cloth. The separated viscous gum is treated with 1% sodium hydroxide then neutralized using acetic acid to prepare the printing paste according to the following recipe:

Natural Dye (powdered)	50 g
Fenugreek Gum	550 g
Urea	150 g
Water	X g
Additive*	Y g
Total	1000g

* Citric acid or Sodium carbonate, or mordant + Citric acid or Sodium carbonate, where: X+Y= 250 gm

3.3. Natural dyes Preparation:

Plant species; (Safflower florets, Henna leaves and Alkanet roots) were dried in dry air at 35° C for 24 hours till the plant materials become crisp. Then, they were ground in laboratory hammer, mill to pass through a 40 mesh (British Standard Screen) and sieve to be very fine powder.

3.4. Study of Printing Factors:

Different factors were studied to determine the best recipe for printing paste to achieve optimum colour shades and higher fastness according to kind of used natural dye and natural fabric.

3.4.1 Effect of Addition of Citric Acid:

The effect of citric acid on printing paste was studied using varied quantities ranged between (without-100 g/kg).

3.4.2 Effect of Addition of Sodium Carbonate:

Variable quantities of sod. carbonate were used i.e (without, 5, 25, 50, 75 and 100) g/kg.

3.4.3 Effect of Addition of Mordants:

Different mordants; (Alum, Iron, Copper and Tannic acid) were used for printing cotton and wool fabrics using the suitable additive i.e. citric acid or sodium carbonate.

3.5. Printing Process:

The cotton and wool fabrics were printed via a manual flat screen technique.

3.6. Fixation Process:

The printed samples were fixed via steaming operation for 7 minutes at 105°C.

3.7. Washing Process:

The fixed printed samples were washed through the following stages:

- Rinsing with cold water then warm water (45°) C.
- Washing with 2 g/L Hostapal CV-ET (non-ionic wetting agent) at 45°C for 15 minutes.
- Rinsing with warm water at 45°C.
- Rinsing with cold water and finally air drying. ^{(5) (16)}

4- Colour strength measurements:

The evaluation of colour strength expressed as K/S of the printed samples was achieved via high reflectance technique. Reflectance measurements of the printed samples were performed on ICS-TEXICON computerised spectrophotometer, model M 520220. These measurements were made in the Textile Printing Department, Faculty of Applied Arts, Helwan University. ⁽³⁾ The colour strength K/S was assessed by applying the Kobelka-Munk equation as follows:

$$K/S = \frac{(1-R)^2}{2R} - \frac{(1-R_0)^2}{2R_0}$$

Where:

R = decimal fraction of reflectance of the printed sample.

R₀ = decimal fraction of reflectance of the unprinted sample.

K = absorption coefficient.

S = scattering coefficient. ⁽²⁾

Results and Discussion

The textile printing with synthetic harmful pastes and dyes affect negatively on the environment. In recent years, there were concerns on the environment

protection. Many attempts were tried to minimize the dangerous from textile field by reduction the used synthetic chemicals. This research was aimed to participate in this target. Using of eco-friendly paste (natural thickener-Fenugreek gum) with natural dyes may be founded as a compatible solution with this aim. Fenugreek seeds gum, which is mainly galactomannan was noticed to be a suitable thickening agent for printing of natural fabrics after treatment with sodium hydroxide and neutralized using acetic acid for controlling the media of paste as neutral media. ⁽¹⁷⁾

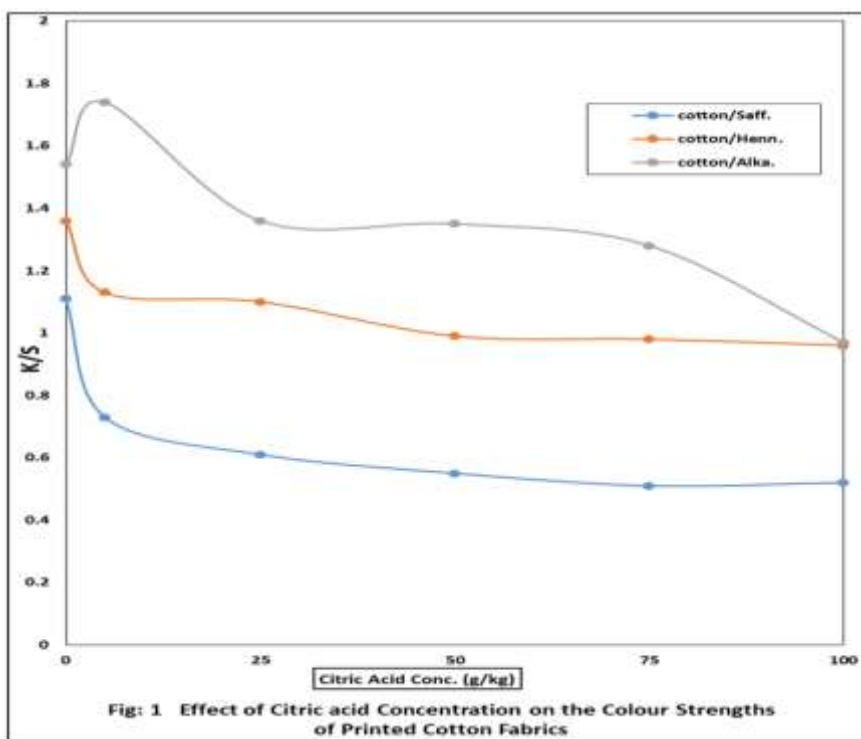
1. Effect of Citric acid Concentration on the Colour Strengths of Printed Fabrics

Cotton and wool samples were printed with the three used plant dyes; Safflower, Henna and Alkanet via various concentrations of citric acid mentioned before under constant printing conditions. The printing paste of Fenugreek gum was neutralized to prevent effect on the pH media of printing of fabrics. The acidity or alkalinity of natural dyes powder when were dissolved in printing paste must be considered. After the printing process finished with all procedures, the colour strengths of the printed samples were measured and the received data are plotted in Figures (1,2).

1.1. Effect of Citric acid Concentration on the Colour Strengths of Cotton Printed Fabrics:

A study of natural dyes printing on cotton fabrics with Fenugreek as thickener was carried out. The colour value results obtained is presented in Fig. (1). It is well observed, that citric acid cannot improve the colour strengths of cotton fabrics printed with Safflower or Henna dyes whereas slightly improvement was observed when printed with Alkanet plant via little quantity of citric acid i.e. (5g/kg).

First of all, cotton fabrics well known has low affinity for natural dyeing or printing without pre-treatments. So it was noticed that K/S values were low generally. Also here, the low K/S values may be attributed to low affinity (printability) of cotton fabrics to print in acidic medium caused by using citric acid that was added to the printing paste. Increasing of citric acid in printing paste resulted in decreasing of dye uptake on cotton fabrics. Also the general acidity media of Safflower and Henna plant which were added in fine powder form to color the pastes may be lowered the affinity to print cotton. Whereas the general alkalinity of alkanet plant powder may be minimize the effect of citric acid and raise the alkalinity of the printing paste which is considered more suitable for cotton fabrics printing.

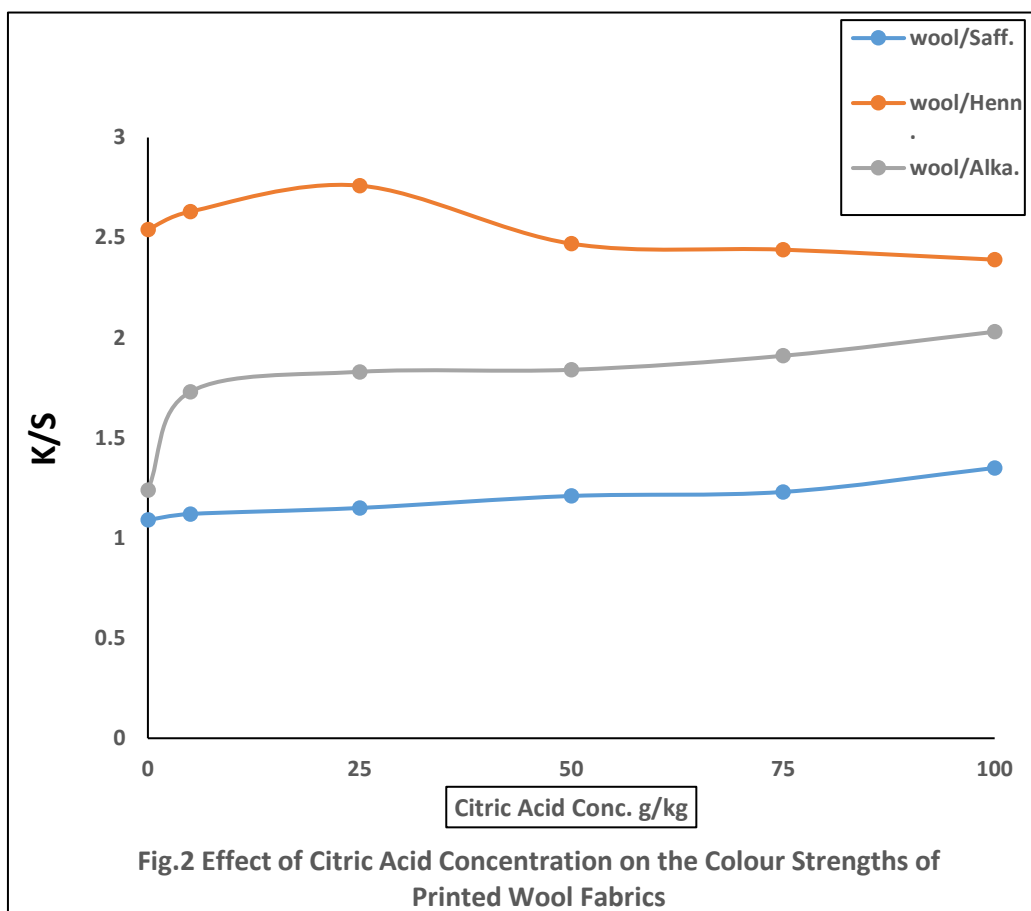


1.2. Effect of Citric acid Concentration on the Colour Strengths of Wool Printed Fabrics:

Reversal behavior was noticed on wool fabrics. The K/S of wool samples printed with Safflower and Alkanet were increased gradually to get the highest value using (100 g/kg) of citric acid and with Henna using (25g/kg) of citric acid. High concentrations of acid may affect negatively on fabrics properties and handling so only appropriate quantities of citric acid are permitted to use.

The natural dye printing process is greatly influenced by variation of the pH of the dye especially in the case of substrates that having ionizable functional groups such as wool fibers. ⁽¹⁰⁾ This aspect may be referred to the ability of wool fibers to attract the different ions and interact with them depends essentially on its degree of ionization, i.e. the number and type of charges on the functional groups.

According to wool behavior, acid media is the most suitable for wool dyeing and/or printing. As shown in Fig. (2), citric acid may be enhanced the acidity of Safflower and Alkanet pastes. In case of Henna plant dye, higher concentrations of citric acid were accompanied with weak colour shades. It may be referred to accumulation of Henna particles in acidic media.

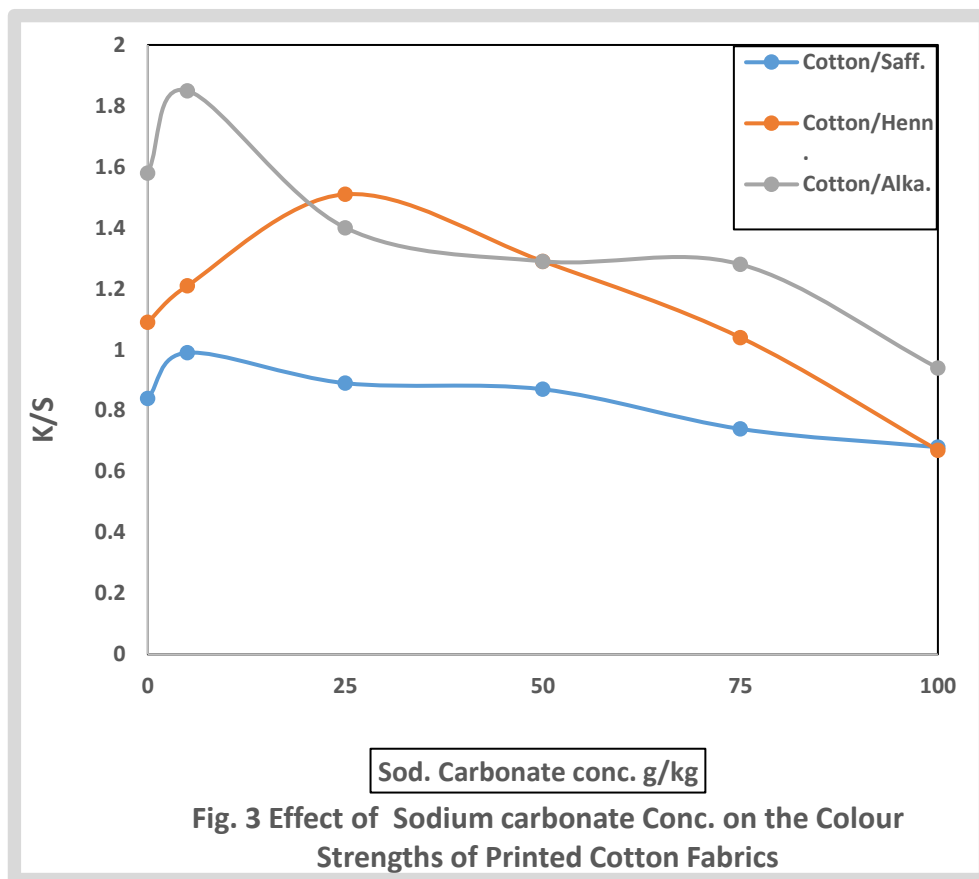


2. Effect of Sodium carbonate Concentration on the Colour Strengths of Printed Fabrics:

In order to optimize the pH of the printing paste, printing were performed with the suitable citric acid or sodium carbonate that were utilized for maintaining the acidic or alkaline medium respectively and the final results (in terms of colour strength) were used for standardization of the pH of the print – paste. Therefore, the relation between the uptake of natural dyes on fibers and the pH of printing pastes depending on kind of fabrics and natural dyes were studied. Fig. (3,4) illustrates the influence of addition of variable values of sodium carbonate in printing pastes on the colour strength of printed fabric.

2.1 Effect of Sodium carbonate Concentration on the Colour Strengths of Cotton Printed Fabrics:

It was clear from the plotted figure that, elevating of pH values using sod. carbonate can caused some remarkable enhancing in the K/S readings for the printed cotton fabrics. Maximum colour strength was attained via 5g/kg Na_2CO_3 in state of printing with pastes contained Safflower and Alkanet dyes. It was cleared from Fig. (1) and Fig (3) that Alkanet dye achieved higher colour strength either using 5 g/kg citric acid or sod. Carbonate. These results may be attributed to the nature of dye, solubility, dispersion and exhaust degree, pH of the paste.



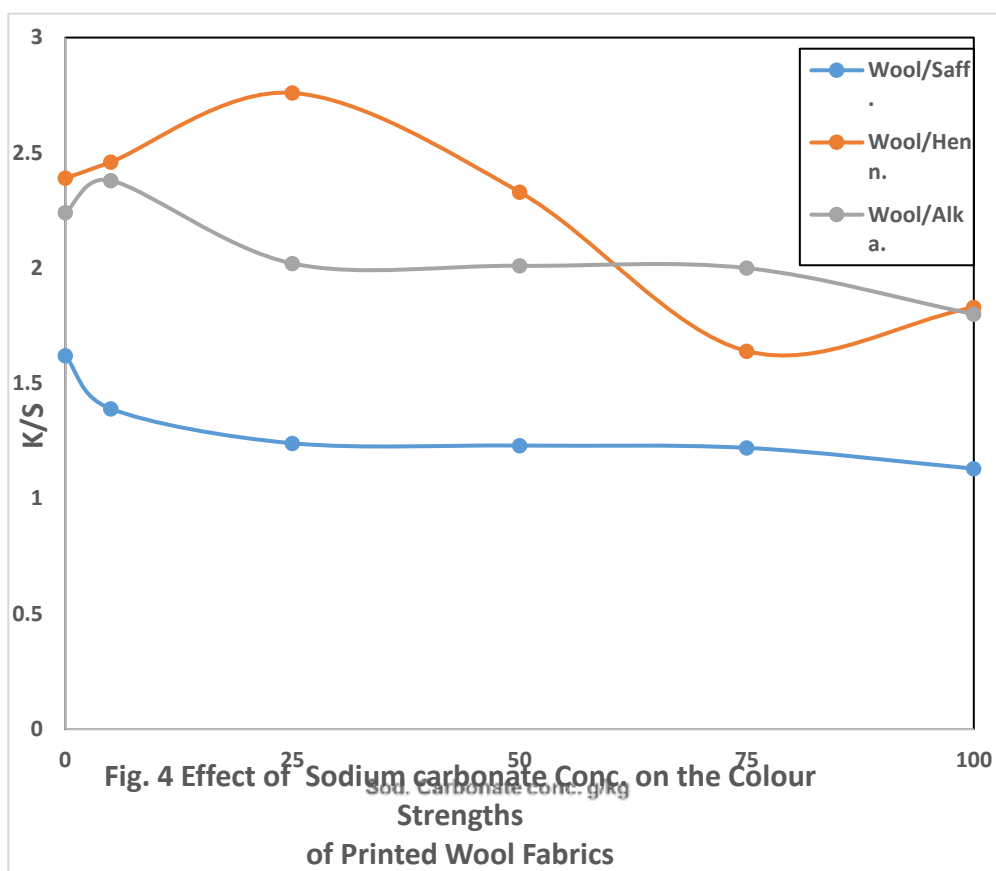
Alkanet was shown ability to give different colour shades according to the media i.e. (red in acidic media blue in alkaline one).

The best colour strength was attained via 25 g/kg Na_2CO_3 in state of printing paste contained Henna dye which means cotton fabrics has better affinity for Henna dye in slightly acid to alkaline media. It is worthy to mention that aspect may be referred by increasing the pH, the solubility of dye increases to be more soluble. More hydroxyl groups may ionize with the formation of negatively charged dye ions resulting in greater enhancement of electrical repulsive forces between them to prevent their aggregation in the printing paste. As a result of dye

disaggregation, the rate of printing will be increased leading to higher colour strengths. ⁽⁴⁾

2.2 Effect of Sodium carbonate Concentration on the Colour Strengths of Wool Printed Fabrics:

From Fig. (4), also there was a noticeable improvement in colour strengths of wool fabrics printed with printing paste contained (25), (5) g/kg with Henna and Alkanet respectively while with Safflower without any colour improvement was noticed.



It can be concluded from the previous figures, that:

- Alkanet has good affinity for cotton and wool fabrics in acid or alkaline media with determine quantities of citric or sod. Carbonate.
- Henna has not achieve better colour strength on cotton fabrics with citric acid though it can achieve good affinity with sod. carbonate also on wool fabrics with measurable acidity and alkalinity.
- Safflower has a good affinity in acid media on wool printing and with low quantity of alkali on cotton printing.
- Fenugreek gum play a successful role as a natural thickening agent in the printing paste for cotton and wool fabrics.

3. Effect of Mordants on the Colour Strengths of Cotton and Wool Printed Fabric:

It has been reported that the mordants are added when using natural dyes to ⁽¹¹⁾

- Increase the fixation of the dyes,
- Obtain a various colour range,
- prevent the fading of dyestuff,
- Deepen, brighten, or dull a colour,
- Improve their overall fastness properties.

Table (1): Effect of Addition of Mordant on the Colour Strengths of Fabrics Printed using Different Natural Dyes

Fabric \ Mordant		Colour Strengths obtained using Different Mordants on Fabrics Dyed with Natural Dyes					
		Safflower		Henna		Alkanet	
		C*	K*	C*	K*	C*	K*
Cotton	without	-	0.88	-	1.19	1.00	1.78
	Alum	-	1.37	-	1.76	0.89	1.18
	Cu	-	2.28	-	2.04	0.98	1.80
	Iron	-	4.25	-	4.89	1.60	4.32
	Tannic Acid	-	0.79	-	1.23	1.07	0.95
Wool	without	2.28	1.09	1.73	1.73	1.32	1.079
	Alum	1.29	0.97	1.50	1.56	1.60	1.63
	Cu	1.28	1.14	1.90	1.55	2.51	1.59
	Iron	1.12	1.96	2.16	3.69	1.37	2.20
	Tannic Acid	1.10	0.86	2.12	1.57	1.70	3.04

C* : Printing with optimum conc. of citric acid.

K* : Printing with optimum conc. of sodium carbonate.

It was obvious from Table (1) that the colour strengths of printed fabrics differ according to the kind of fabrics, dyes, printing media, and used mordants. The mordants have the greatest effect on the resulted K/S were ordered as:

* Iron > Cu > Alum > Tannic acid.

- * In case of addition of sod. carbonate in printing of cotton fabrics with three plant dyes.
- ** Iron > Tannic acid > Cu > Alum.
- ** In case of addition of citric acid in printing of cotton fabrics with three plant dyes.

It can be concluded that Iron was the most effective mordant on cotton fabrics with all used natural dyes which can be attributed to its role in increasing the redness and darkness of the colour shades. Also Iron was the recommended mordant in mordanting of wool fabric printed with Safflower and Henna dyes.

Other cleared notes were reported from mordanted samples without readings:

- Tannic acid shifts the colour to brown.
- Alum brightens the colour shades.
- Iron dulls and darkens the colour and shift it into grey tones.
- Copper darkens the dye colour and shifts it to green.

4 Conclusion

Utilization of fenugreek gum as a thickening agent success in printing paste using natural dyes which contribute in minimizing pollution problems in the environment. Generally speaking, it is clear in most cases the K/S of the protein fabrics (i.e. wool) is relatively higher than cellulosic fabrics (i.e. cotton). This is expected since the tannins present in the natural dyes are high molecular weights compounds containing phenolic hydroxyl groups that enable them to form effective crosslinks between proteins; where they form three types of bonds:

1. Hydrogen Bond: it is formed between phenolic hydroxyl groups of the tannins and free amino and amido groups of the proteins.
2. Ionic Bond: it is formed between suitably charged anionic groups and tannin groups on the protein.
3. Covalent Bond: it is formed via the interaction of any quinone or semiquinone group present in tannins with the suitable reactive groups in the proteins. ^{(17) (6)}

However, in cellulosic fabric (i.e. cotton) the tannins could form only two types of bounds:

1. Hydrogen Bond: which formed between hydroxyl groups of cellulose and the phenolic hydroxyl groups of the tannins.
2. Covalent Bond: which formed via the interaction of quinone or semiquinone group present in tannins with the suitable reactive groups in the cellulose. ^{(10) (12)}

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