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Ultraviolet Protection of Cotton Fabrics and Their Blends Using Natural Plant Extracts by Printing Style

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THE purpose of the present study was to harness the natural plant extracts to print and treatment the cotton fabrics and its blends. This study reports the impart Ultraviolet protection factor (UPF) to woven fabrics such as cotton and its blend which printed with colourants of Melia azedarach extract. The results showed a difference in the functional groups between the printed and unprinted samples, which indicates that the new functional groups are specific to the treatment materials used in printing paste . The results also indicated that the UPF values for the printed samples are much higher than unprinted one, and some of these values are higher than the specifications limit for the UPF tests. Also results showed that the K/S values of printed samples are higher than unprinted samples, irrespective of the nature of the fabric used especially in the case of samples treated with citric acid as mordant in which it shows a direct increase directly proportional to the increased concentration. The results also indicated that the air permeability of the printed samples is lower than that of the unprinted samples as a result of the treatment operations. While under investigation Colour fastness to rubbing, washing, perspiration and light showed values were very good to excellent In all samples and at all concentrations .

Keywords: Printed cotton and its blend, Melia azedarach, Tannin, Citric acid, UPF, FTIR and K/S.

Introduction

The importance of sunlight is that it is the main source of energy, the wavelength range of Sun radiation has about 0.7 nm to 3000 nm and the effective spectrum reaching on the surface of earth spans from 290 nm to 3000 [1] .

UV radiation amounts to about 6% of solar radiation [2] . The ultraviolet solar radiations (UVR) divides to three regions: UV-A (320 to 400 nm), UV-B (290 to 320 nm), and UV-C (200 to 290 nm). UV-C is fully absorbed by the atmosphere and does not reach to the earth surface . Although UV-A light is the predominant component of terrestrial UV radiation and rather weak in the carcinogenesis, excessive exposure to UV-A light causes aging and wrinkling of the skin [3] . Also, UV-A causes little visible reaction on the skin but has been shown to decrease the immunological response of skin cells [4]. UV-B is most responsible for the development of skin cancers result in skin damage such as sunburn, premature skin ageing and allergies, particularly in white-skinned people .

Small doses of ultraviolet (UV) solar radiation are beneficial to humans [5] . for example human skin generates vitamin D when it is exposed to small doses of UV radiation. but overdose of UV radiation causes dangerous [6], So it is fortunately, the higher-energetic part of Solar radiation is filtered off by the stratosphere and does not reach the Earth's surface [7].

Therefore The increasing trend in the production of fabrics to protect against UV radiation is the result of increased health risks resulting from exposure to this radiation [8] caused by The depletion of ozone in the upper earth's atmosphere [9].

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Nature and its plants and herbs are a source of medicinal materials since ancient times [10].The majority of the country's developing population and nearly 25 percent of the country's developed population are using herbal medicines for treatment and disease prevention [11].

For example, Melia azedarach is one of the plants that possess several medicinal properties, such as antioxidant, anti-fungal, anti-bacterial, anti-helminthic and anti-malarial activity[12]. Also dry powder fruit of Melia azedarach was used for the treatment of stomach, diabetes and fever[13]. It is one of the most useful plants in traditional medicine as well as modern medicine where leaves are used in treatment anthelmintic, leprosy and scrofula [14] .

In the current study, we used the ethanolic extract of Melia azedarach plant to print and treat Cotton fabrics and its blend to raise the protection efficiency against the sun's ultraviolet rays . Also, the extract was used as a natural pigment colour in the printing paste.

Material and Methods

Fabrics

1- 100% bleaching cotton fabric(Giza 86, Plain weave1/1,144gm/m^2, Number of threads in the warp 84, Number of threads in the weft 66, count number yarn of warp 24/1, count number yarn of weft 24/1) supplied by Misr company for spinning and weaving, Mehalla El-Kubra, Egypt

2- 100% bleaching blending cotton fabric(45% poly ester – 55% cotton, Plain weave1/1, 116 gm/m^2, Number of threads in the warp 111, Number of threads in the weft 68, count number yarn of warp 36/1, count number yarn of weft 36/1) supplied by Misr company for spinning and weaving, Mehalla El-Kubra, Egypt .

Mordants

Tannic acid ($C_{76}H_{52}O_{46}$) [Loba chemie pvt.ltd , India].- Citric acid ($C_6H_8O_7$)

Plant material

Ripe fruits of the plant were collected from Al Qanater alkhairia Nurseries, Egypt, in April 2018. The ripe fruits of M. azedarach were washed well many times with tap water to get rid of impurities and dust and then dried in shade at temperature between 21-38^oC for 20 days, These fruits were then grinded to obtain finely grounded dried powder, using a special laboratory mill (ARC - Department of Fiber Chemistry) because the fruits are very solid .

Extraction procedure

The dried powder plant materials(50 gm) were extracted via maceration in 1000 ml ethanol (50%) and left for 72 at room temperature With shaking In dark around bottom flask to avoid oxidative factors After this period it is left for three hours at 70 ° C using the water bath then the mother solution is filtered using filter paper and vacuum pump then the residual plant material refluxed again with ethanol (50%) , these process repeated four times until reaching 0 in mass loss . finally In the end, extract was evaporated using water bath to obtain a dry weight from Melia azedarach extract . the curd extract preserved at 4 ° C Until it is used .

Printing technique

All the pastes were applied to the fabrics through flat screen by traditional technique

Thickening agent

High viscosity Carboxyl Methyl Tamarind 8 (CMT), which is Anionic thickening agent and it has Viscosity of 8% paste from 33000 — 37000 cps at 250C 6/20 Brookfield viscometer RVT model 9 -Il in 8% solution

Dye Materials

Melia azedarach extract used as a pigment colour with concentration (50%, 75% and 100%).

Preparation of printing paste

The pastes used for application of dye extract (Melia azedarach as a pigment colour in printing were prepared using the following recipe :-

Melia Azedarach extract ---------	400g
Thickener----------------------------	80 g
Water -------------------------------	520 g
Total	1000g

Mordanting of fabrics

Mordanting of the fabrics (cotton and cotton/ polyester) were conducted utilizing pre-mordant technique using a concentration of 1% tannic acid at 80 ° C for 30 min . In the same way, citric acid was used as mordant, as it was used at a concentration of 1% at 45 °C for 30 min.

Fixation

After printing, the printed samples were dried at room temperature, which was approximately 30 ° C for 18 hours. Then the samples were subjected to fixation by steaming at 120 ^{O}C for 30 min **.**

Washing

After the fixation process, the printed samples were washed to remove the excess material from the surface of the fabric and to ensure the stability of the printed materials . The washing process was done in four steps :

1. Rinse the samples gently using cold water
2. Rinse samples using hot water
3. Rinse the samples using non-ionic soap 2 g / L at 45 ° C for 20 minutes.
4. Finally, the samples are rinsed with cold water.

The washed samples are left to dry at room temperature.

Methodology

FTIR Analysis

Fourier Transform Infrared Spectroscopy (FTIR) of the printed and unprinted fabrics were implemented by using an FTIR spectrophotometer (Nicolet 380 by Thermo Fisher Scientific) in the region of 400-4000 cm-1 with a spectral resolution of 4 cm-1

Colour strength (K/S) .

The colour strength of the printed samples expressed as K/S was assessment conducted by (spectrophotometer optimatch 3100® from the SDL Company.)
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Measurement of UPF for printed fabrics

This test was carried out using a device Spectrophotometer according to the International Standard AATCC Test Method 183Transmittance or Blocking of UV Radiation through Fabric.

Colour fastness Evaluation

Colour fastness for washing

This test has been conducted in accordance with ISO 105 – C10: 2015 «Textiles -- Tests for colour fastness -- Part C10: Colour fastness to washing with soap or soap and soda «

Colour fastness for perspiration

Colour fastness perspiration tests (acid and alkaline) were carried out according to international standard ISO 105-E04: 2018 « Textiles -- Tests for colour fastness -- Part E04: Colour fastness to perspiration» .

2.12.4.3. Colour fastness to light

This test was carried out on printed samples in accordance with ISO 105-B01:2014 " Textiles -- Tests for colour fastness -- Part B01: Colour fastness to light: Daylight " .

Colour fastness to rubbing

This test was carried out on printed samples in accordance with ISO 105-X16:2016 "Textiles -- Tests for colour fastness -- Part X16: Colour fastness to rubbing -- Small areas" .

Air permeability (AP)

This test was carried out on printed samples using FX3300 (Textest, Switzerland) with a pressure applied of 196 Pa, according to ASTM D737- 04, and five samples were tested in each case .

Result and Discussion

FTIR Analysis

All the printed and unprinted fabric were analyzed by FT-IR in order to characterize the new polymeric structures.

Comparing the results with the control samples (unprinted samples) , several new bands appeared in the printed samples which related with flavonoids and phenolic compound in Melia azedarach extract .

Fig. 1 depicts the FTIR spectrums of unprinted cotton fabrics. The unprinted cotton fabric possessed functional groups expressed by the following peaks: The wide band which showed around band at 3000–3500 cm^{-1}, associated with the stretching vibration of –OH. The next high memorable peaks was 2907 cm^{-1} which corresponds to The C–H stretching vibration of cellulose . while the band at 1104 cm^{-1} linked with the C-O-C stretching vibration [15,16]. Other notable peaks were founded in the area 1313, 1157, 1020 cm^{-1} are Resulting from bending and stretching vibrations of C–H, C–O, COO which existent in the cellulose respectively[17].

The spectra printed cotton fabrics (Fig.3) showed the characteristic strong peak at 3400–3000 cm^{-1} for broad stretching band the –OH of polysaccharide structure of Melia azedarach .

On the other hand, Fig. 2 showed the FTIR spectrums of unprinted cotton /polyester fabrics . the FTIR spectrums of unprinted cotton /polyester shows the presence of the following peak : 3986.34, 3301.70, 2852.34, 1710.63, 1407.85, 1276.71, 1236.21, 1045.28, 1014.42, 1002.85, 721.28, 663.43 cm^{-1}. the band at 3986.34 and 3301.70, 2852.34 cm^{-1} corresponds to (O-H) Hydrogen bonded Alcohols and (C-H) alkanes respectively[18]. The band at 1710.63 cm^{-1} corresponds to carbonyl group (C=O) stretching Which specializes in polyester fibers without cotton fibers[19].

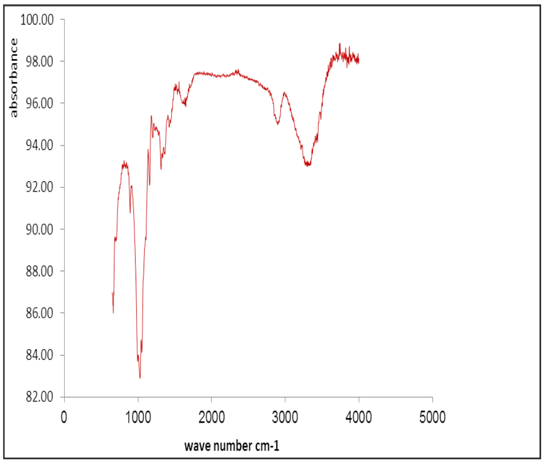

Fig . 1. FTIR of printed cotton samples

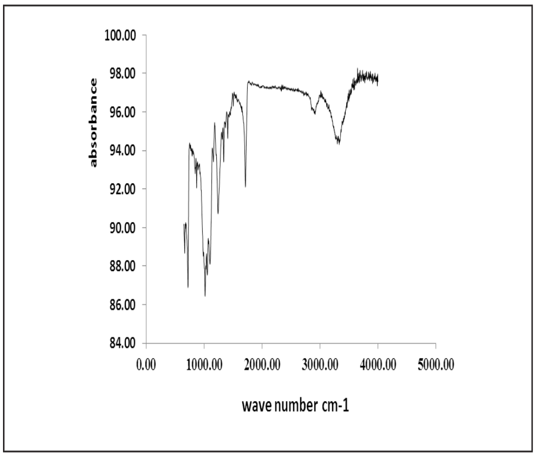

Fig . 2. FTIR of unprinted cotton/polyester Samples.

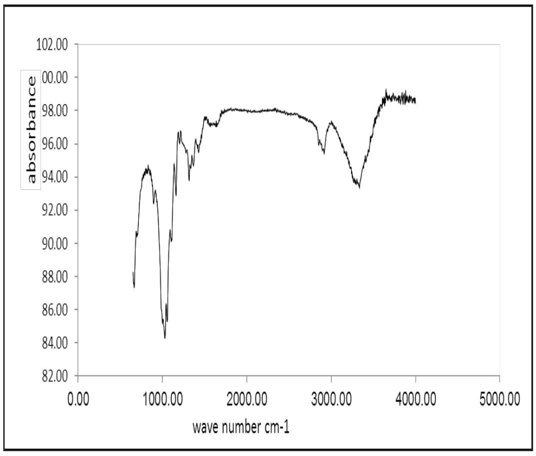

Fig . 3. FTIR of unprinted cotton samples .

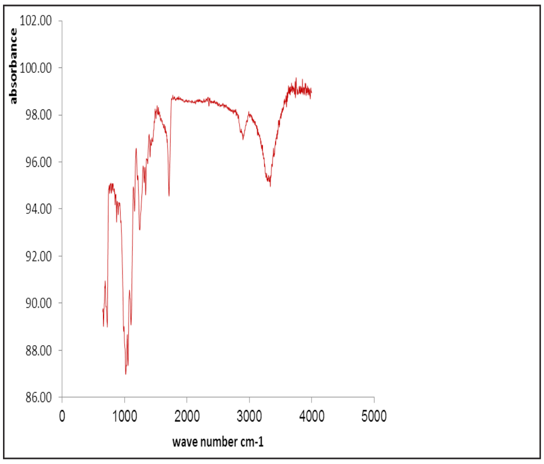

Fig . 4. FTIR of printed cotton/polyester Samples.

It was shown in Fig. 4. The FTIR spectrums of printed cotton /polyester have revealed the presence of various chemical constituents with various peaks values such as peak at 3400–3000 cm^{-1} for broad stretching band the –OH of polysaccharide structure of Melia azedarach, But not the same in pure cotton samples . This can be explained by the fact that the polyester fibers are weakly bound to the active substances in the M.azedarach extract, unlike the pure cotton samples.

UPF for printed fabric

Ultraviolet protection factor' (UPF) is the scientific term which used to indicate the amount of ultraviolet protection provided to skin by fabric [20] .From table 1, it could be seen the values of UPF "Ultraviolet Protection Factor" for printed and unprinted fabrics .Also the result indicated to the fabrics mordanting with tannic acid and citric acid. From the data obtained and shown in Table 1, the data mean there is a great difference in the UPF properties between printed and unprinted fabrics. Whereas the UPF of unprinted cotton and cotton/polyester fabrics were 1.78 and 2.79 respectively . It can be said that there is no protection from ultraviolet sunlight or it is absent . the results in table 1 reveal that the excellent UPF capacity of printed cotton and cotton /polyester fabrics especially cotton samples, particularly those treated with citric acid as a mordant . The results are as follows:

The UPF of samples which treated with citric acid gave the following results : cotton sample gave 82.10 . While cotton/polyester gave 73.54. the samples treated with tannic acid gave the following results: the cotton samples gave 52.27, 46.24 and 42.04 at concentration 100%, 75% and 50% respectively . Similarly the cotton/polyester samples gave 48.62, 38.78 and34.68 at concentration 100%, 75% and 50% respectively.

From the previous results, it was found that the cotton samples were higher in the UPF than the cotton / polyester samples. Likewise, the samples treated with citric acid as mordant were higher than the samples treated with tannic acid as mordant, and it was also observed that with increasing the concentration, the protection factor of the printed samples increases. The high UPF for the printed samples alike from unprinted samples can be explained by the fact that the M.azedarach extract which used in the printing process contains many organic compounds such as flavonoids and phenols ... etc, which in turn absorb the harmful ultraviolet rays and convert them into non-thermal energy Harmful . Also, The increase in colour strength is offset by an increase in the UPF for the fabrics, and this is confirmed by the K/S test, as the samples treated with citric acid have a higher colour strength(K/S), which is also the highest in the protection factor against the sun's ultraviolet rays(UPF).

Colour strength (K/S)

One of the important tests is the colour intensity test, It is denoted by the symbol K/S, Where K = Absorption coefficient and S = Scattering coefficient)The colours are given in CIELAB coordinate system: L* presents luminosity, a* presents redness-greenness (+value=red, -value=green), b* presents yellowness-blueness (+value = yellow, -value = blue) [21].

After completing all the printing stages, the printed samples were beige to brown .According to Table 2, the Colour data (L*, a*, b* and ΔE) revealed that, L* values of printed fabric and mordanting with tannic acid decreased than those in case of mordanting fabrics with citric acid and fabrics. This means that the dark shade is achieved and accepted with K/S values. Likewise A decrease of a* and b* values mean a less reddish and yellowish shade respectively. The ΔE values were obtained in case of citric acid higher than in case of tannic acid. The auxochromic groups (OH and COOH) in phenolic compounds in *Melia azedarach* extract were able to complex compounds formation, depending on these group number suitable for forming complexes with mordant and combined with the fabric via hydrogen bonding.

Table 2 shows the effect of M.azedarach concentrations on the colouration properties of the printed fabrics . It is clear from the data of Table (2) that the K/S is directly related to the M.azedarach concentrations Decreasing the M.azedarach concentrations is accompanied by great loss in colour strength. The maximum colour strength (K/S) value was obtained at concentration 100%, 75% and 50% respectively for the both type of fabrics .

From the table 2, also It is obvious that K/S value of the printed cotton fabrics is higher than printed cotton/polyester fabric Where the highest value was 2.1563 and 1.9242 for cotton and cotton /polyester respectively . This may be due to number of active group such hydroxyl group And amorphous regions on cotton fabric surface more than on cotton/ polyester fabric surface, So cellulosic fabrics i.e. cotton acquire high affinity for natural colours than blended or Synthetic fibers i.e. polyester. It has been reported that natural dyes are high molecular weight compounds containing phenolic hydroxyl groups which enable them to form effective cross-links, where they form two types of bonds namely. (a) Hydrogen bond: which is formed between the hydroxyl groups of cellulose and phenolic hydroxyl groups of natural dye. (b) Covalent bond: it is formed by interaction of any semi Quinone or Quinone group present in natural dye with any suitable reactive groups in the cellulose[22].

TABLE 1. Ultraviolet protection factor (UPF) of printed fabric.

No	Type of fabric	Conc.	UPF (control samples)	UPF(T)	UPF(C)
1	cotton 100%	50%	-	42.04	-
2	cotton/polyester		-	34.68	-
3	cotton 100%	75%	-	46.24	-
4	cotton/polyester		-	38.78	-
5	cotton 100%	100%	-	52.27	82.10
6	cotton/polyester		-	48.62	73.54
7	unprinted cotton	-	1.78	-	-
8	Unprinted cotton/polyester	-	2.79	-	-

(T) samples which mordant with Tannic acid ,(C) samples which mordant with Citric acid .

Irrespective of the fabric used; the K/S values of samples which pre-treatment with citric acid are higher than samples which pre-treatment with tannic acid at the same concentration.

Air permeability

There are many factors that affect the comfort properties of fabrics, such as air permeability, thermal insulation, permeability of steam and water, heat transmittance, and fabric structure. The air permeability feature is a very important comfort feature for fabrics and clothing it Maintains thermal balance for its wearer [23]. It can be defined as “ the flow rate of air passing vertically through a particular unit area of the fabric by measuring a certain pressure difference of air across the fabric test area through a specific time period “[24].

Table. 3 shows the effect of two pre-treatments (tannic acid and citric acid) as mordanting and M.azedarach extract as a finishing agent on the air permeability for both fabric typ. It can be observed that both of the treatments cause the reduction of air permeability for both fabric where the air permeability rate for the unprinted samples was 349 $l/m^2/s$ and 423 $l/m^2/s$ for cotton and cotton/polyester fabrics respectively. While the maximum air permeability of the both printed fabrics was 165 and 215 $l/m^2/s$ at concentration of 100% from M.azedarach extract using tannic acid as a mordant. In order to explain this, it is necessary to know the most important factors that affect the air permeability of fabrics, which are the porosity of the fabrics As the more porosity, the more air permeability and vice versa. As a result of the printing process and crosslinking the structural and superficial changes occurred in the fabrics, which were to reduce the amount voids and pores in the fabrics where the air passes through it, which led to a decrease in the air permeability.

TABLE 2. Colour specifications and K/S value of printed and unprinted samples.

Mordant type	Conc. ME	Fabric type	K/S	L*	a*	b*	ΔE
Tannic acid	50%	cotton 100%	1.1586	-17.72	1.46	20.52	27.15
		cotton/polyester	0.8573	-17.53	1.21	19.72	26.41
	75 %	cotton 100%	1.2417	-18.84	2.01	21.46	28.63
		cotton/polyester	0.8958	-18.30	1.20	19.38	26.68
	100%	cotton 100%	1.3685	-19.88	1.75	21.40	29.26
		cotton/polyester	0.9014	-17.46	1.20	19.45	26.16
Citric acid	100%	cotton 100%	2.1563	-31.49	2.73	23.06	39.13
		cotton/polyester	1.9242	-33.97	2.23	21.80	40.42
Control sample	-	Cotton 100%	0.6166	-	-	-	-
Control sample	-	cotton/polyester	0.8068	-	-	-	-

TABLE 3. The Air permeability of printed and unprinted fabrics.

No	Description	concentration	Type of fabric	*Air permeability* (Unprinted fabrics)	*Air permeability* (T)	*Air permeability* (C)
1	Printed fabrics	50%	100% cotton	-	129 $l/m^2/s$	-
2			blending	-	172 $l/m^2/s$	-
3		75%	100% cotton	-	139 $l/m^2/s$	-
4			blending	-	181 $l/m^2/s$	-
5		100%	100% cotton	-	165 $l/m^2/s$	34 $l/m^2/s$
6			blending	-	215 $l/m^2/s$	95 $l/m^2/s$
--	Unprinted fabrics	-	100%Cotton	349 $l/m^2/s$	-	-
8		-	blending	423 $l/m^2/s$	-	-

(T) samples which mordant with Tannic acid , (C) samples which mordant with Citric acid.

It is also clear that the fabric samples which treated with citric acid is more effective in reducing air permeability than treated with tannic acid This can be explained by the fact that in the case of citric acid treatment, there is significant interaction and crosslinking between components of the printing past and the surface of the fabric, which led to a greater decrease in the number of pores present in the fabric and consequently a greater decrease in air permeability than the samples treated with tannic acid.

Colour fastness Evaluation

The Table 4 shows the various fastness categories (washing, alkali perspiration, acid perspiration) of printing fabrics using M.azedarach with natural mordant .

Colour Fastness defined as " resistance power of colour of a textile material against a lot of factors during production and usage[25] **.**

Colour fastness for washing

Colour fastness to washing is the widespread characteristic parameter, which is one of the most important characteristics of fabrics and clothing from the viewpoint of consumers. This test determines the change &loss of colour in the washing process by a consumer and the behavior of staining of lighter or other garments that may be washed with it**.** From the results in table 4 it is observed that the printed fabrics which mordanting with tannic acid and citric acid show good wash fastness where the colour change was found from 4 to 4-5 and staining showed 4 to 4-5. This is due to the formation of hydrogen bonds between mordent, M.azedarach extract and fabric, which gave this high stability[26].

Colour fastness for perspiration

Colour fastness for perspiration is an important test for fabrics, especially those directly adjacent to skin cells According to the table 4 the colour fastness of printed samples with different M.azedarach concentrations showed excellent perspiration fastness Whether acid or alkali , as well as whether in the colour change of printed fabrics or staining in adjacent fabrics where the colour change was found 4 to 4-5 in acidic perspiration solution and 4 to 4-5 in alkaline perspiration solution, Likewise in colour staining showed 4 to 4-5 for acid and 4 for alkaline. This confirms the high stability and bonding strength between mordant, M.azedarach extract and the surface of fabrics .

TABLE 4 . The value of colour fastness(washing,rubbing, perpiration and light) of printed samples.

Fabric type	**Conc.**	**mordant type**	**Washing fastness**		**Rubbing fastness**		**Perspiration fastness**				**Light fastness**
							Acidic		**Alkaline**		
			Alt.	**St**	**dry**	**wet**	**Alt.**	**St**	**Alt.**	**St**	
Cotton 100%	50%	Tannic acid	4-5	4-5	4-5	3- 4	4-5	4-5	4-5	4	6
	75%		4-5	4-5	4-5	4-5	4-5	4-5	4-5	4	6
	100%		4-5	4-5	3-4	3	4-5	4-5	4-5	4	6
Blending	50%		4-5	4-5	4	4	4-5	4-5	4-5	4	6
	75%		4-5	4	4-5	4	4-5	4-5	4-5	4	6
	100%		4-5	4	4	3	4-5	4-5	4-5	4	6
Cotton 100%	50%	Citric acid	4	4	4-5	4-5	4-5	4-5	4-5	4	6
	75%		4	4	4-5	4-5	4	4	4	4	6
	100%		4	4	3-4	3	4	4	4	4	6
Blending	50%		4	4	4	4	4	4	4	4	6
	75%		4	4	4-5	4	4	4	4	4	6
	100%		4	4	4	3	4	4	4	4	6

Colour fastness to light

All coloured materials have susceptibility to fading due to light, as the highest persistence of light indicates that these pigments absorbed the lengths that they do not reflect again, because light is energy and when this energy is absorbed and reflected again, damage to the pigments and colours may occur[27].Also Light fastness of printed fabric is influenced by physical state and concentration of dye, chemical, nature of the fibers and mordant type[28].According to the table 4 the light fastness of printed samples with different M.azedarach concentrations showed excellent light fastness. Which confirms that dyes(*M. azedarach*) are stable to photo degradation and the interaction of printing paste is good.

Colour fastness to rubbing

Colour fastness to rubbing is an important test for measuring colour fastness as it is checked the colour transferred from the coloured textile surface to other surface by rubbing in wet and dry condition.

The Results of the colour fastness to crocking are reported in Table 4. Where the results indicated a high colour fastness to rubbing for printed fabrics which printed with different concentration of M.azedarach extract and mordanting with tannic acid and citric acid where the results were from 3-4 to 4-5 for dry rubbing while in the case of wet rubbing the results values were from 3 to 4-5 .

This shows that majority of dye molecules is fixed well on fibers and surface residual Dye molecules is minimal. Also It may be the result that dye molecules formed intermolecular hydrogen bonding with the mordant and the fabric.

Conclusion

Protecting people from the sun's harmful UV rays has become an imperative, especially because of the ozone hole and the ever-increasing incidence of skin cancer. The textiles are considered one of the ways to protect against harmful ultraviolet radiation, so it was necessary to raise the protective factors for it .The results confirm the possible use of some plant extracts(*M. azedarach)* to improve the UV protection properties of woven fabrics such as cotton and cotton/polyester by printing style . The results also showed a significant decrease in the rate of air permeability in all printed samples, Also the colour fastness properties in case of screen printed samples i.e. washing, perspiration, light and rubbing for all above the data found to be very good to excellent.

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