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Application of Novel Eco-friendly Natural Dye Extracted from Leaves of Neem on Silk Fabric

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Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

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ABSTRACT

In the present work, optimization of extraction natural dye from neem (*Azadirachta indica*) leaves with respect to dye bath concentration on silk fabric was studied before and after exposing to artificial daylight for 160 h. The effect of different dye bath concentrations in the range 40-240 g/L on the reflectance spectra of the unexposed and exposed silk fabric was followed by using spectrophotometric technique. The absorption coefficient, optical band gap energy and extinction coefficient as well as color strength were calculated from the reflectance data and the wash fastness was tested. The color parameters: the relative brightness (L*), color constants (a* and b*), whiteness index (W), color difference (ΔE), chroma (C*), hue (H), and tint (T) and the CIE tristimulus values (x_r, y_r, and z_r) were also determined. The obtained results showed that the absorption and extinction coefficients, color strength and color parameters were highly affected by changing dye concentration. In addition, it was also noticed that the variations in the values of the optical band gap energy with increasing dye concentration may be due to the induced structural change in the system. In addition, the dyed silk samples have shown good washing fastness properties. The washing fastness grade for color change ranged from 4 to 5, whereas from 4 to 4/5 for color staining with all samples. The present study indicates that the natural

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dye extracted from neem leaves has good potential in textiles dyeing and meet the environmental future demands technology of high quality fantastic dyed pattern through an economical point of view.

Keywords: Azadirachta indica; neem plant; natural dye; silk fabrics; textiles dyeing; optical properties.

1. INTRODUCTION

Silkworm silk is a beautiful lustrous fiber produced naturally. Silk fiber consists of two biomacromolecules; fibroin (about 75%) and sericin (about 25%) by weight; which has been used as a premium textile material [1]. Silk fiber, also, consists of a filamentous protein, a non-filamentous protein and other impurities such as pigments, wax, carbohydrates and inorganic salts. Silk is a natural protein composed of 18 amino acids with various reactive functional groups [2]. Silk has gentle luster, smooth and soft texture, good drapability, good hygroscopicity and excellent comfortability [3]. Sericin strengthens the silk fiber, but it is necessary to degum before dyeing because it makes lack luster [4-6]. Silk is a biocompatible [2,4] biodegradable material [7] with low inflammatory reaction in the body [6] and with superior mechanical properties [8]. Silkworm silk can be used as a suitable biomaterial in tissue engineering and in clinic surgery as stitches besides its application in fabrics. Silk fiber has several weaknesses such as poor antibacterial activity, poor UV protection performance, deterioration, yellowing and wrinkling which during usage exerts negative appearance [9]. Silk fiber has a general chemical formula [NH_2.CHR.COOH]. It is bonded to dyes due to ionic interaction between the free chemical groups of dyes and the carboxyl groups of silk. The most important groups for dyeing of silk are COOH and NH_2 [7]. In dyeing process, the dye solution is absorbed and diffused into the fiber.

Textile dyeing processing industry is one of the significant environmental polluters by using synthetic dyes in comparison with natural dyes [10,11]. Thus renewed international interests have arisen in natural stains due to the increased awareness of the environmental and are safer in use with minimum health hazards [11,12]. The natural colors present in plants and animals are pigmentary molecules which impart color to the materials [13]. They may have a wide range of shades and can be obtained from various parts of plants including roots, bark, leaves, flowers, and fruit [14]. Furthermore, natural dyes are known to exhibit better biodegradability, less toxicity, eco-friendly alternative to synthetic dyes and some stains also possess medical properties [15,16]. Moreover, naturally dyed textiles have therapeutic properties provides relief for arthritis, diabetes, headaches and over-excited nerves and is also suitable for blood circulation [17]. Natural dyes are applied as a green and sustainable dyeing process due to their biocompatibility, low toxicity, abundant availability, antibacterial activity, deodorizing performance, UV protection, and eco-friendliness [18,19].

Azadirachta indica commonly known as "Neem", "Nimtree" and "Indian Lilac" belongs to Meliaceae botanical family and being abundantly available [20]. *Azadirachta indica* is one of two species in the genus Azadirachta, and is native to India and the Indian subcontinent. The images in Fig. 1 illustrate the neem tree, tender neem leaves used as medicine, neem flowers and neem fruits.

Neem is a fast-growing tree that can reach a height of 15–20 m and rarely 35–40 m. The branches are wide and spreading. The fairly dense crown is roundish and may reach a diameter of 15–20 meters in old, free-standing specimens. Neem tree can grow in many different types of soil, but it thrives best on well drained deep and sandy soils. The opposite, pinnate leaves are 20–40 cm long with 20 to 31 medium to dark green leaflets about 3–8 cm long. White and fragrant flowers were arranged in drooping axillary panicles of 25 cm long. The inflorescences belong to the third degree which bears 150-250 flowers of 5–6 mm long and 8–11 mm wide, each. In the same individual tree; protandrous, bisexual flowers and male flowers existed. The fruit is a smooth and has different shapes from elongate oval to nearly roundish, and when ripe is 1.4–2.8 cm by 1.0–1.5 cm.

The principal constituents of neem leave include; protein (7.1%), carbohydrates (22.9%), minerals, calcium, phosphorus, vitamin C, carotene, etc. [20,21]. An extract from the leaves can be

Fig. 1. Neem tree, tender neem leaves used as medicine, neem flowers and neem fruits

prepared as an alcoholic tincture or as tea, and this alcohol extract has a dark green color [22,23]. The chemical composition of the extract contains glutamic acid, tyrosine, aspartic acid, alanine, praline, glutamine and cystine like amino acids, and several fatty acids (dodecanoic, tetradecanoic, elcosanoic, etc.). But physically the neem leaves have a pleasant odor and plant pigments like chlorophyll mainly responsible for green color. The *Azadirachta indica* extract is reportedly officious against a variety of skin diseases, septic sources, and infected burns. The most important quality of these compounds is that they are less toxic to warm-blooded animals like a human. Thus, considering its less toxicity and effectiveness against microorganism, it is expected to be one of the safest and most effective colorants cum antimicrobial agent for textiles [24,25].

In the present work, optimization of extraction natural dye from neem (*Azadirachta indica*) leaves concerning dye bath concentration on silk fabric was studied before and after exposing to artificial daylight for 160 h. The effect of 6 different dye bath concentrations (40, 80, 120, 160, 200 and 240 g/L) on the reflectance spectra of the unexposed and exposed silk fabric was followed by using the spectrophotometric technique. The color parameters (L*, a*, b*, W, ΔE*, C*, H, and T) and the CIE tristimulus values (x_r, y_r, and z_r), were determined. The absorption coefficient, optical band gap energy, and extinction coefficient, as well as color strength, were also calculated.

2. EXPERIMENTAL DETAILS

2.1 Materials

2.1.1 <u>Fabric samples and extraction of colorant</u>

In the present investigation, white silk fabrics of 55 g/m^2 in weight and 0.06 cm in thickness produced in Akhmim, Egypt, was used without any purification. Dried leaves of neem plant were used for obtaining the neem dye without the application of any mordant in the dyeing process. 50 g of neem leaves were immersed after crushing in 500 mL of distilled water and allowed to boil for one h. It was mentioned that, the dyeing process was done without the application of any mordant.

A Shimadzu (VIS) Double Beam Spectrophotometer with standard illuminant C (1174.83) model V-530 and band width 2.0 nm in the range 200-700 nm with accuracy ±0.05% was used to evaluate the optical density of the extract solution after filtration. The determination of the optical density was taken as a measure of concentration. The absorption spectrum of the aqueous extract of the neem leaves was shown in Fig. 2. From the figure, three peaks were presented: the first peak at UV range has λ_{max} = 320 nm of optical density 3.765; the second peak

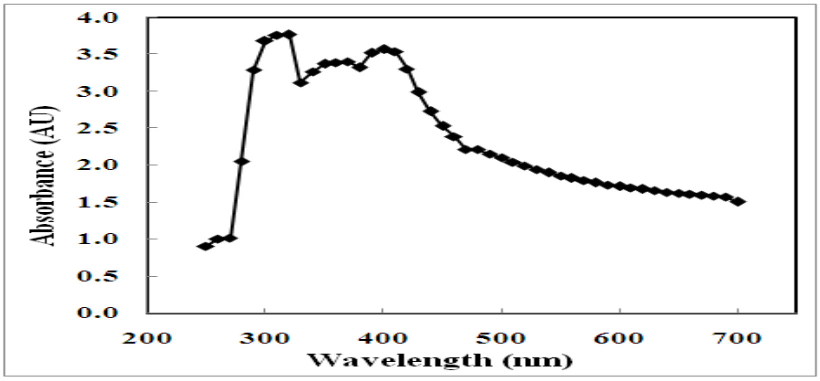

Fig. 2. The absorption spectrum of the aqueous extract of the neem leaves

at λ_{max} = 370 nm of optical density 3.402; and the third one at visible region has λ_{max} = 400 nm of optical density 3.569. The dried leaves of neem plant were found to discharge color in hot water very easily. Increasing the number of leaves from 20 to 120 g per 1000 mL water boiled for one h was accompanied by the increase in color strength and depth in color (easily seen by naked eye). The extracted dye (for all concentrations) was exposed to artificial daylight for 160 h and then examined spectrophotometry. The absorption spectrum of these aqueous extract of neem leaves shows three peaks at the same positions as that presented in Fig. 2.

2.1.2 Dyeing method

Silk fabrics were dyed by using a liquor ratio of 1:50 at pH = 5, at temperature 60ºC for 45 minutes for each concentration [26]. The samples were thoroughly washed with cold and then dried at ambient temperature. Different shades were obtained by using various levels (40, 80, 120, 160, 200 and 240 g/L) ranging from hell yellow to dull yellow.

2.1.3 Reflection measurements

The measurements in the visible region from 400-700 nm for the unexposed and exposed undyed and dyed silk fabrics with different concentrations of neem dye was carried out using a Shimadzu (VIS) Double Beam Spectrophotometer with standard illuminant C (1174.83) model V-530 and band width 2.0 nm covers the range 200-2500 nm with accuracy ±0.05%.

The effect of different neem dye bath concentrations on the absorption coefficient, optical band gap energy, extinction coefficient and color strength of the dyed silk fabrics have been determined. From the reflectance spectra, the absorption coefficient (α), extinction coefficient (K) and the color strength (α/S) were calculated in the visible range covered from 400-700 nm by using the following equations [27-32].

$$\alpha = (1/d)\ \ell n\ [(1-R)^2/T] \quad (1)$$

$$K = \alpha\lambda/4\pi \quad (2)$$

$$\alpha/S = (1-R)^2/2R \quad (3)$$

Where R is the reflectance, T is the transmittance (≈ 10^{-3}), and d is the thickness of the sample (about 0.06 cm). α represents the absorption coefficient (is dependent on the dye stuff) and S is the scattering coefficient (is dependent on the substrate). On the other hand, the color properties were analyzed using the CIE Colorimetric System, CIE 1931 2-degree Standard Observer. The tristimulus values (x_r, y_r, and z_r), the relative brightness (L*), color constants (a* and b*), whiteness index (W), color difference (ΔE), chroma (C*), hue (H) and tint (T) were performed [33-37].

All the silk fabrics under test were exposed to artificial daylight using Tera Light Fastness Tester [38] for 160 h at a temperature of 25 ± 2°C and at a relative humidity of 65 ± 5%. A standard blue scale was hanged alongside the samples (ISO 105-B02).

2.1.4 Determination of fastness properties

The silk samples were washed as stated by the conditions mentioned in AATCC Test Method 61-2009 to determine the change in color and staining of adjacent fabrics after cleaning. A specimen of the textile in contact with one or two specified adjacent fabrics was mechanically agitated under specified conditions of time 30 minutes and at 50°C in a soap solution without a fluorescent whitening agent (WOB), then rinse and dried. The rating scale of washing fastness for color change was from 1 (very poor), 2 (poor), 3 (fair), 4 (good) to 5 (excellent). This was assessed concerning the original fabrics using the gerg scale.

3. RESULTS AND DISCUSSION

3.1 Absorption Coefficient, Extinction Coefficient, and Color Strength

The reflectance percentages (R%) as function of wavelength in the visible range 400-700 nm for undyed as well as unexposed (a) and exposed (b) neem dyed silk fabrics with different dye bath concentrations (40, 80, 120, 160, 200 and 240 g/L) were shown in Fig. 3. It is clear from Fig. 3a. for the unexposed dyed silk fabrics that, R% values increase markedly with increasing wavelength for all samples and decrease gradually with increasing the dye bath concentration up to 240 g/L in comparison with the undyed sample. These observed variations indicate that there is a high color change as a function of wavelength by increasing neem concentration. Similar behaviors and trends with more differences were observed for all samples after exposing to artificial daylight for 160 h (Fig. 3b).

The absorption coefficients (α) of the undyed as well as the unexposed (a) and exposed (b) silk fabrics dyed with different concentrations were calculated in the visible wavelength range 400-700 nm (i.e., photon energy range 3.10-1.77 eV) by using Equation 1 from the reflectance spectra (Fig. 3.) and were represented as functions of wavelength and photon energy in Figs. 4 and 5, respectively. It is clear from the figures that remarkable decrease in the absorption coefficient values with increasing the wavelength for all samples. Also, α values increase with increasing the concentration of neem through the whole wavelength and photon energy ranges for both the unexposed and exposed samples to artificial daylight for 160 h.

As reported previously [39-41], the change in the absorption coefficient may be due to the difference in the chemical bonds between the fabric and the dye which form other molecular species and leads to the formation of new color centers. The detected increase in absorption coefficient values either by increasing the neem concentration and exposure to the artificial daylight may be attributed to the change in the molecular configuration of the fabric which may indicate to the formation of new color centers. Also, the data indicates that dyeing with neem and exposure with daylight leads to rupture of the bonds and formation of free radicals. These observed variations may be due to modification in molecular structure introduced as a result of the degradation process. Also, the dye components role was to strength the linkage between the reactive species of the silk fabric chemical groups and their polar groups.

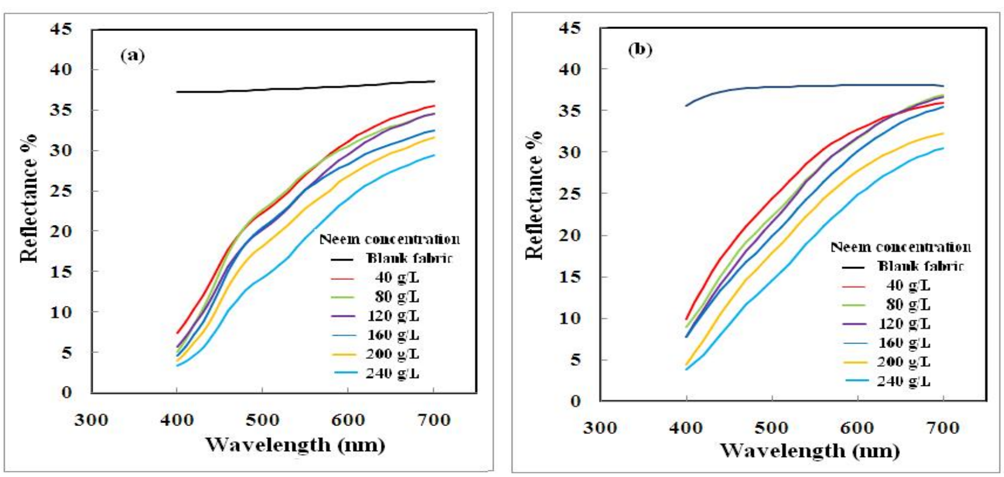

Fig. 3. The reflectance % spectra of undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h as functions of wavelength in the visible region

The energy represents the width of the tail localized states in the normally forbidden band gap was given by applying Urbach relation as [27-30]:

$$\alpha = \alpha_o \exp [h\upsilon/E_b] \quad (4)$$

Where α_o (= constant) is the absorption coefficient at hv = 0, υ is the frequency of radiation, and E_b is the band tail energy value. Fig. 6 shows the dependence of ℓn α on hv for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h. It is clear from the figure that, each curve could not be represented by straight lines relation which means that the absorption does not follow the quadratic relation for inter-band transitions to verify Urbach rule [30]. On other hand, as observed from Fig. 6b that the variation between ℓn α and hv are more regular with increasing the neem concentration for the exposed samples but still showed complicated behaviors which may due to the change in the total number available states caused by exposing to the artificial light according to the compromise between the degradation and/or cross-linking processes [42,43]. Also, the observed changes may be due to the variation in the internal fields due to the interaction between neem and silk fabrics chemical groups.

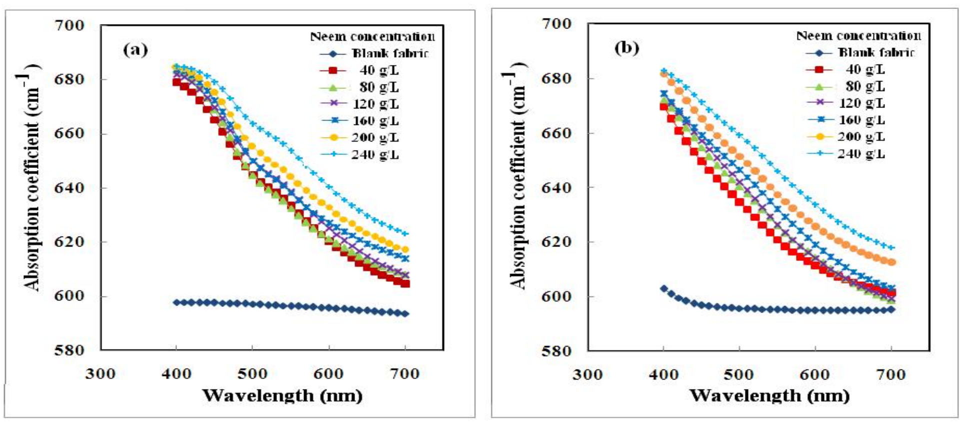

Fig. 4. Plots of the absorption coefficient values of undyed as well as neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h against wavelength

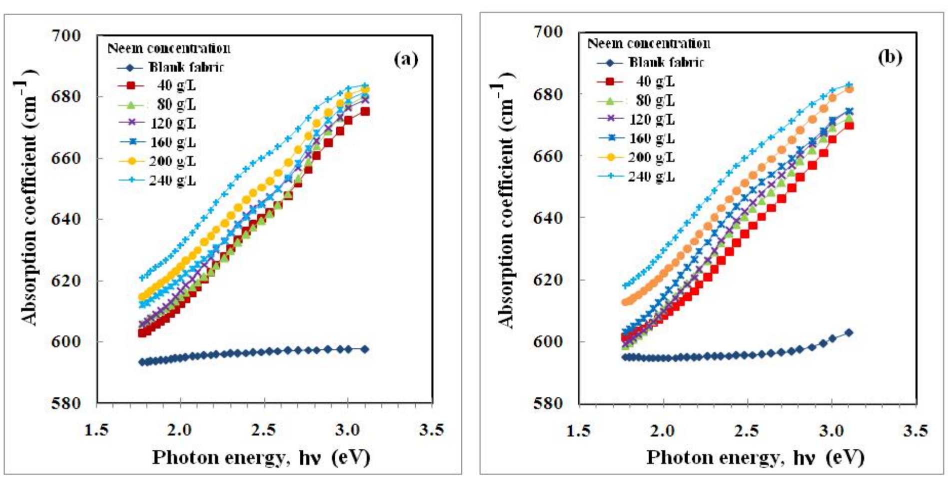

Fig. 5. Plots of the absorption coefficient values of undyed as well as neem silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h against photon energy (hv)

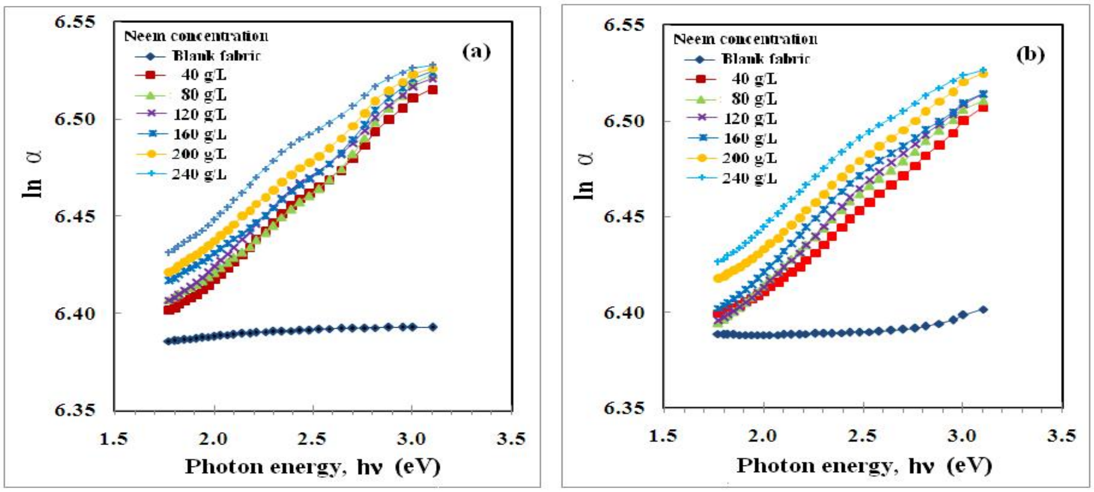

Fig. 6. Variation of ℓn α with hν for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h

According to Tauc's model and from the following relation, the power part obeys the Tauc was given as [30,31]:

$$\alpha h\upsilon = B\,(h\upsilon - E_g)^n \qquad (5)$$

Where B is the band tail parameter in the range from 10^5 to 10^6 $(cm.eV)^{-1}$ which represents the slope of Tauc's edge and n is the electronic transition equals ½ or 2 for allowed direct or indirect transitions, respectively. Fig. 7 shows the variations of $(\alpha h\nu)^2$ with hν for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h. From the figure, the values of allowed direct energy gap (E_d) were calculated by extending the linear parts of the curves to zero absorption and were tabulated in Table 1. It is observed from the table that, firstly, the E_d values increase with increasing the dye bath concentration up to 80 g/L and then decrease gradually for the unexposed samples. The E_d values of the fabrics exposed to the artificial light show the same behavior with increasing the dye bath concentration. These increases indicate that the benefits of E_d show the dependence on the composite and creation of localized states in the band gap. Moreover, the obtained increase in the optical energy gap (E_d) may be attributed to the change in molecular configuration which leads to rupture of the bonds and formation of free radicals and then structural changes were occurred [27,41-44].

Fig. 8. shows the dependence of $(\alpha h\upsilon)^{1/2}$ on the photon energy (hυ) for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight. Again, as mentioned before, the obtained changes may be due to the change in molecular configuration which leads to structural variations.

The extinction coefficient (K) characterized the photonic material and represented the properties of the material to light and can be calculated by using Equation 2 [29]. The variations in the extinction coefficients (K) as functions of wavelength for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h were illustrated in Fig. 9. It was noticed from the figure that, the variation behaviors of K for all samples were similar through the whole wavelength range. Also, the values of the extinction coefficient were found to be small (in the order 10^{-4}). This indicates that silk fabrics may be considered as an insulating material at room temperature [45]. Moreover, the increase in the extinction coefficient values with increasing the concentration of neem dye without or with exposure to artificial light shows that the fraction of light lost could be due to scattering.

By determining the color strength (α/S) value of the dyed material, a close relationship to the amount of dye absorbed by the fabric was obtained. Using Equation 3, the effect of increase in the neem dye bath concentration on the color strength of the non-dyed for the unexposed (a) and exposed (b) silk fabrics was illustrated in Fig. 10. It is clear from the figure that the values of the color strength decrease sharply with increasing wavelength for all samples. On the other hand, α/S values increase with increasing the number of neem leaves in the extraction bath up to 240 g/L for both unexposed and exposed samples through the whole wavelength region. This means that, higher the concentration of neem higher is the coloring component extracted and higher is the color strength of the fabric dyed with these extracts which means that the deepest shade was occurred.

Also, the effect of the dye bath concentration can be attributed to the correlation between dye and silk fibers. Since neem dye is a water-soluble dye and containing anionic groups, these groups would interact ionically with the protonated terminal amino groups of silk fabrics through ion exchange reaction. Then, due to this ionic attraction, the dye-ability of the fiber would increase with increasing the neem dye concentration. This increase in the dye-ability may be due to: 1) the enhanced desorption of the dye, 2) the greater availability of the dye molecules in the vicinity of the fiber, and 3) the increase in diffusion due to the rise in the amorphousity of the fiber. The present data are in good agreement with the previously reported results [46-48].

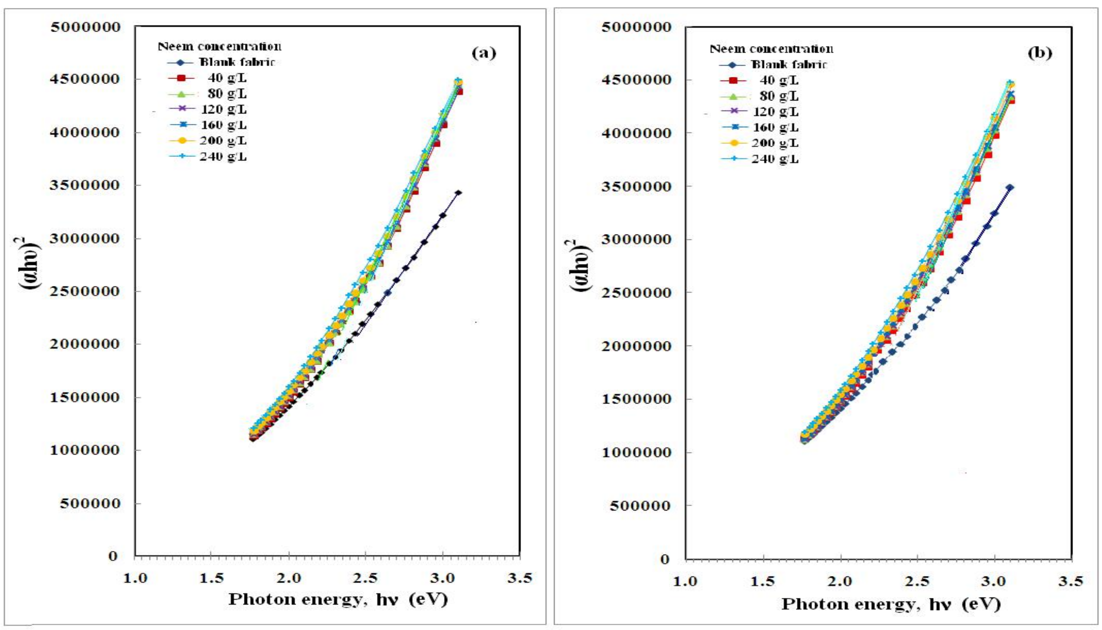

Fig. 7. Variations of $(\alpha h\upsilon)^2$ with photon energy (hv) for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160h

Table 1. Values of the direct energy gap (E_d) for undyed and neem dyed silk fabrics with different neem concentrations before and after exposure to artificial daylight for 160 h

Concentration of neem (g/L)	Direct energy gap (E_d) (eV)	
	Before exposure	After exposure
Undyed fabric	1.481	1.604
40	1.746	1.784
80	1.773	1.740
120	1.726	1.742
160	1.723	1.719
200	1.689	1.726
240	1.621	1.652

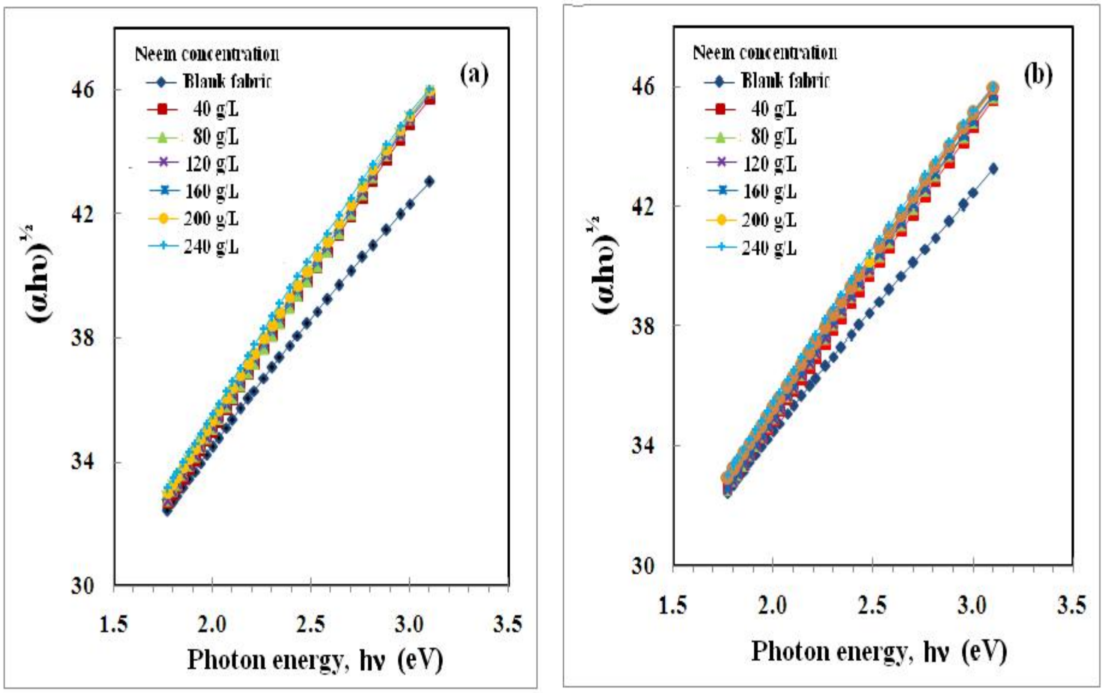

Fig. 8. Variations of $(\alpha h\upsilon)^{1/2}$ with photon energy (hv) for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160h

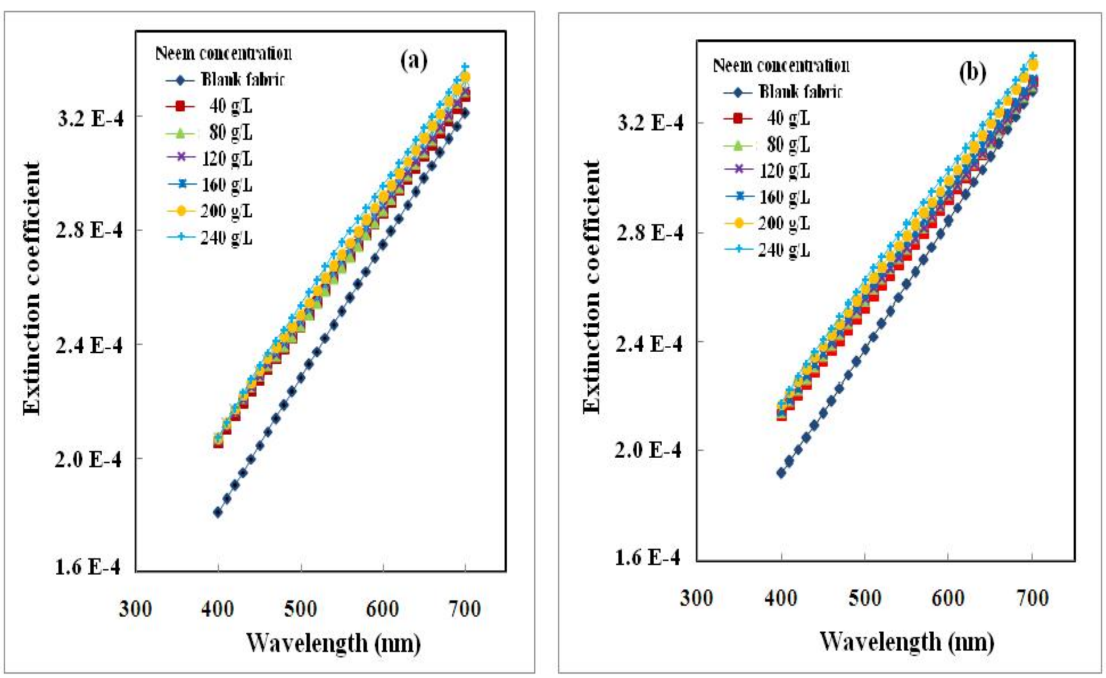

Fig. 9. Dependence of the extinction coefficient on wavelength for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h

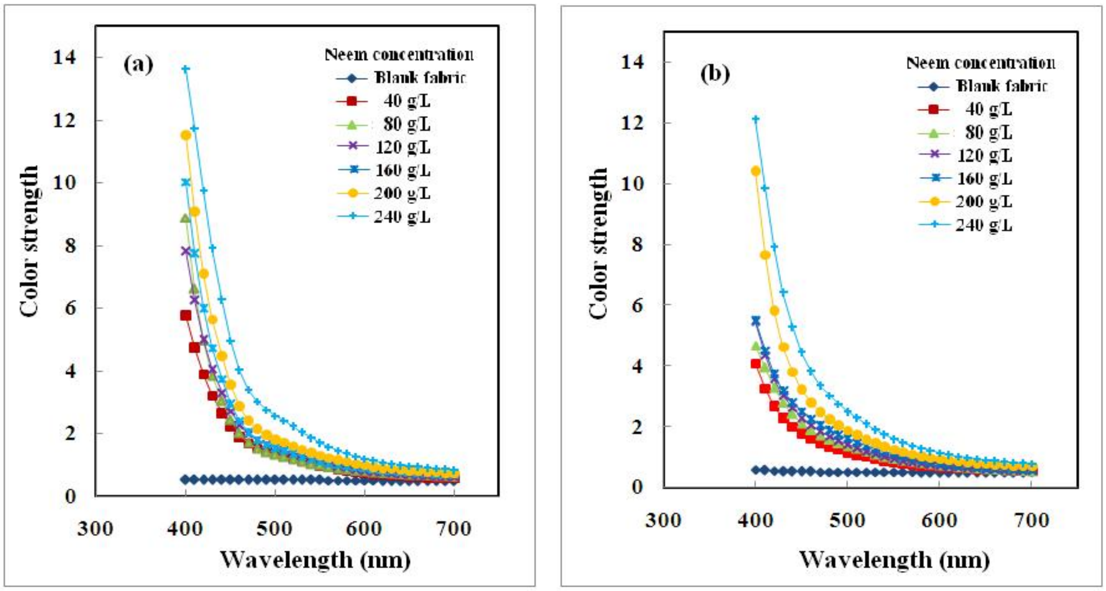

Fig. 10. Dependence of the color strength on wavelength for undyed and neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h

3.2 Tristimulus Values and Color Parameters

The variations of the tristimulus values (x_r, y_r, and Z_r), the relative brightness (L*), color constants (a* and b*), whiteness index (W), color difference (ΔE), chroma (C*), hue (H), and tint T) for undyed and neem dyed silk fabrics with different dye bath concentrations (40, 80, 120, 160, 200 and 240 g/L) before and after exposure to artificial daylight for 160 h were calculated from the values of the reflectance percentage spectra represented in Fig. 3. The tristimulus reflectance values (x_r, y_r, and z_r) for the undyed silk fabric as well as the unexposed (a) and exposed (b) neem dyed silk fabrics with different dye bath concentrations were plotted against wavelength (400-700 nm) and were illustrated in Figs. 11, 12, 13 and 14, respectively. It was observed from the figures that, the behaviors of x_r, y_r, and z_r for the samples are similar, and there is no change in the peak positions either without or with exposure to the artificial daylight.

Table 2 illustrates the maximum tristimulus reflectance values at their peak positions for undyed and neem dyed silk fabrics under investigation. It was noticed from the table that, the tristimulus values decrease with increasing the neem dye bath concentration. Also, the tristimulus values of the exposed silk fabrics to artificial daylight were higher than those of the unexposed values for all samples. Also, the x_r, y_r, and z_r values for all samples either unexposed or exposed to artificial daylight were lower than that of the undyed silk fabric.

The color properties such as: the relative brightness (L*), color constants (a* and b*), whiteness index (W), color difference (ΔE), chroma (C*), hue (H), and tint (T) were analyzed using the CIE Colorimetric System, CIE 1931 2-degree Standard Observer and were tabulated in Table 3. From the table it was noticed that: The relative brightness, L*, shows a decrease in their values with increasing neem concentration and exposure to artificial daylight which means that the fabric becomes fader in color. The values of the color constant, a*, increase by increasing neem concentration and exposure to artificial daylight which indicates that, there is an increase in red component instead of green component. The values of the color constant, b*, increase by increasing neem concentration and exposure to artificial daylight which indicates that there in an increase in yellow component instead of blue one. The detection of the whiteness index (W) and tint, T, values indicated opposite behavior in comparison with the color constants, a* and b*. The observed decrease in the whiteness index, W, values and the results of the color scales, C* and H, as well as color difference, ΔE, indicated that variations in color difference between the fabrics and the neem dyed were occurred due to the presence of different neem concentration and/or exposure to artificial daylight.

As noticed from Table 3, the observed changes in the values of the color parameters by increasing neem concentration and/or exposure to artificial daylight for 160 h may be due to the change in the physical bonds and then changes in the molecular configuration of the fabric which may lead to formation of new color centers. Also, the obtained results of the color parameters were of great importance for the improvement of the optical properties of the fabrics.

3.3 Wash Fastness Test

Table 4 shows the wash fastness properties of the undyed and neem dyed silk fabrics with different dye bath concentrations. It was noticed that as the dye concentration increase, the samples give an excellent rating. Other samples give rating good to very good. This is because the ionic bonds between dye molecules and the carboxyl groups in the silk fiber are strong. Also, in this natural dye, there are chemical groups such as C=C and C=O which adsorb in the fiber and increase washing fastness due to its saturation properties.

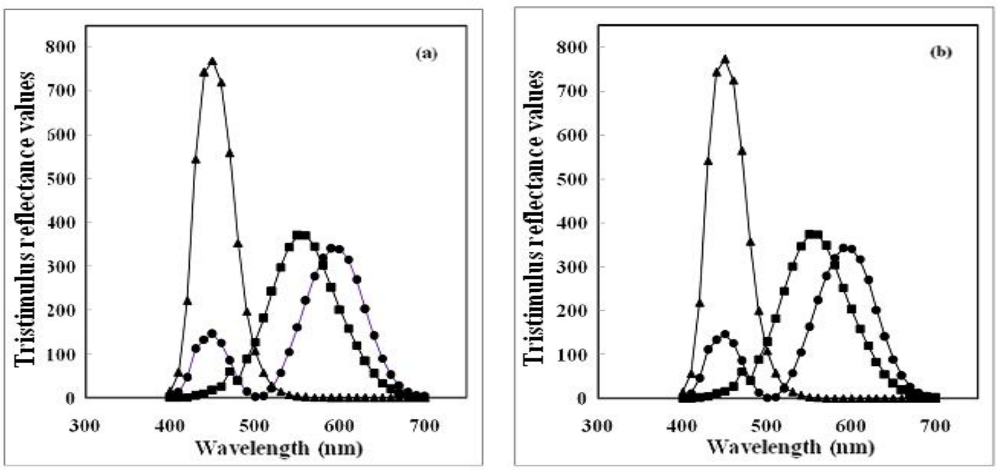

Fig. 11. Variations of the tristimulus reflectance values [x_r (▲), y_r (■) and z_r (●)] with wavelength for undyed silk fabrics before (a) and after (b) exposure to artificial daylight for 160 h

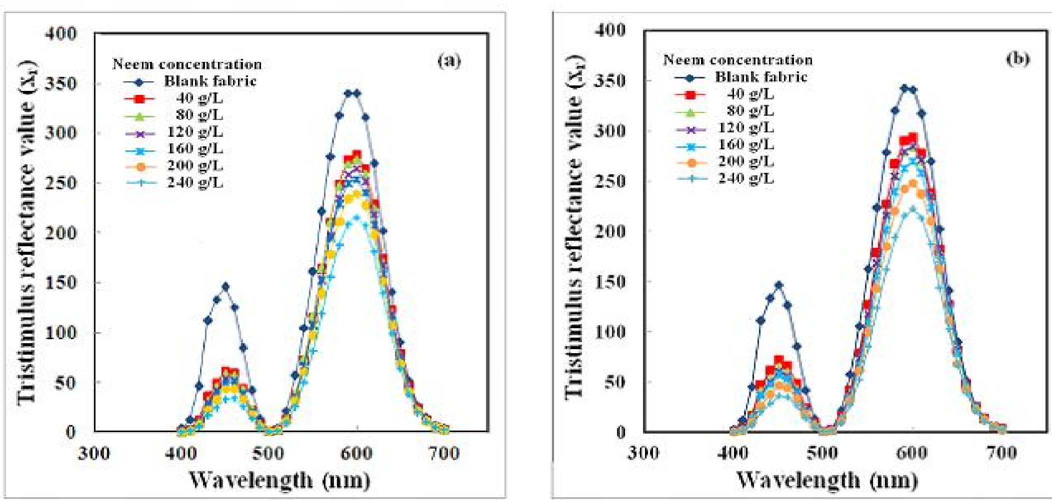

Fig. 12. Variation of the tristimulus reflectance value (x_r) with wavelength for neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h

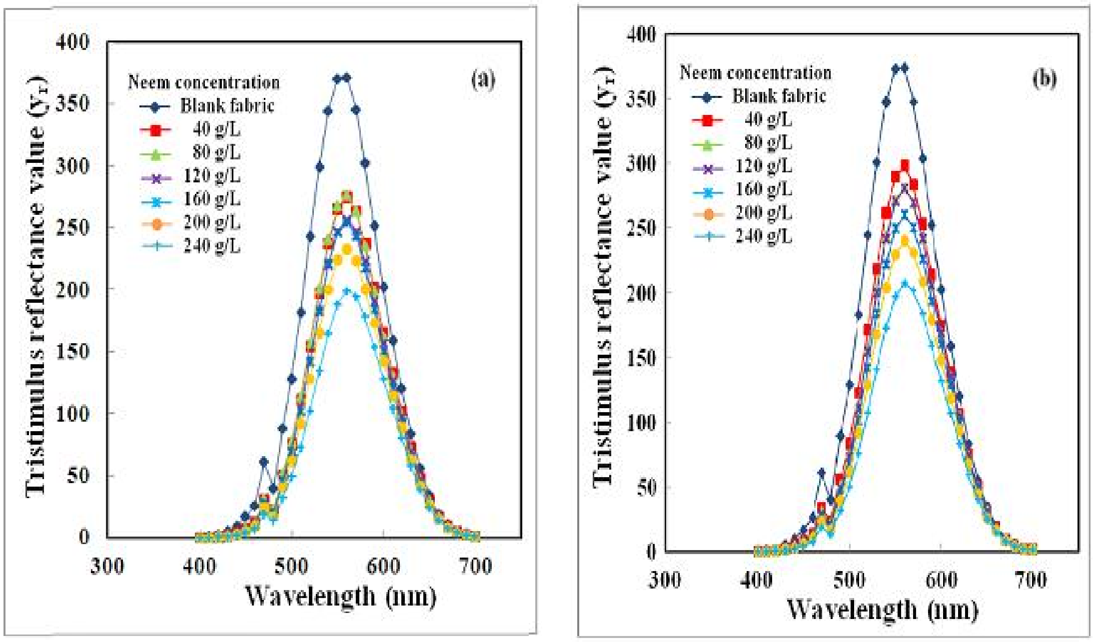

Fig. 13. Variation of the tristimulus reflectance value (y_r) with wavelength for neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h

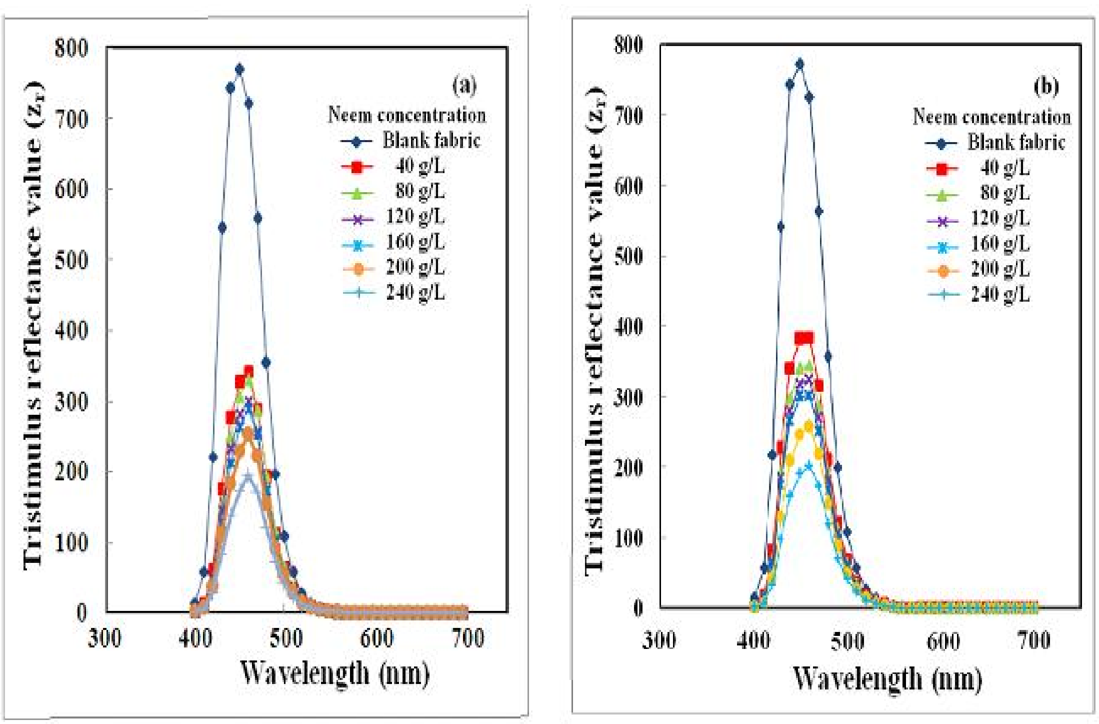

Fig. 14. Variation of the tristimulus reflectance value (z_r) with wavelength for neem dyed silk fabrics with different dye bath concentrations before (a) and after (b) exposure to artificial daylight for 160 h

Table 2. The maximum tristimulus reflectance values (x_r, y_r, and z_r) at their peak positions for undyed and neem dyed silk fabrics with different dye bath concentrations before and after exposure to artificial daylight for 160 h

Concentration of neem (g/L)	x_r		y_r	z_r
	λ = 450 nm	λ = 600 nm	λ = 560 nm	λ = 460 nm
Unexposed silk fabric samples				
Undyed fabric	146.0691	339.8218	371.4584	743.6189
40	62.0946	279.0685	274.9741	342.0297
80	58.1611	273.4286	270.1697	330.7398
120	53.5936	264.8124	257.2465	300.4856
160	50.1397	254.0206	254.8922	289.2546
200	43.3988	239.9007	232.9487	252.2351
240	33.0628	215.7222	199.1904	194.2606
Exposed silk fabric samples to artificial daylight for 160 h				
Undyed fabric	146.6637	340.8764	373.810	773.141
40	72.5489	292.7307	298.1331	382.4428
80	64.5259	283.5043	281.8621	340.7334
120	60.6367	284.5782	280.7478	319.0635
160	56.9633	269.5439	259.9992	300.2829
200	46.8626	248.1558	239.6284	247.0369
240	36.2294	222.0694	207.3794	190.9841

Table 3. The color parameters for undyed as well as neem dyed silk fabrics with different dye bath concentrations before and after exposure to artificial daylight for 160 h

Color parameters	Concentration of neem (g/L)						
	0	40	80	120	160	200	240
L*							
Before exposure	75.75	58.74	57.00	58.71	56.63	54.58	50.85
After exposure	67.98	60.88	58.99	59.22	57.23	55.07	51.67
a*							
Before exposure	-0.09	2.30	2.82	2.43	2.69	2.83	4.86
After exposure	0.15	1.50	3.09	2.86	4.17	3.39	5.06
b*							
Before exposure	-0.86	20.29	22.28	22.28	23.46	24.43	26.23
After exposure	0.74	17.99	21.58	19.77	20.76	23.62	25.50
W							
Before exposure	-	-107.06	-125.20	-121.50	-135.30	-147.30	-169.60
After exposure	33.50	-87.90	-114.7	-102.6	-113.9	-139.5	-161.80
ΔE[1)]	-	22.95	25.52	24.69	26.59	28.43	32.16
ΔE[2)]	1.44	3.11	2.12	3.53	4.45	1.66	1.11
ΔE[3)]	-	4.16	2.78	4.66	5.45	2.66	2.66
C*							
Before exposure	0.80	20.42	22.46	22.29	23.47	24.51	26.68
After exposure	0.80	18.05	21.80	19.98	21.17	23.76	26.00
H							
Before exposure	101.50	83.52	82.80	88.90	88.31	85.26	79.50
After exposure	101.46	85.23	81.85	81.77	78.64	81.83	78.78
T							
Before exposure		-17.70	-20.70	-13.50	-15.30	-20.40	-31.90
After exposure	0.00	-13.70	-20.50	-18.80	-23.70	-23.90	-31.70

1) *Variation in ΔE* due to different neem dye bath concentrations*
2) *Variation in ΔE* due to exposure to artificial daylight for 160 h*
3) *Variation in ΔE* due to due to different neem dye bath concentrations after exposure to artificial daylight for 160 h*

Table 4. Wash fastness rating for undyed as well as neem dyed silk fabrics with different dye bath concentrations

The concentration of neem (g/L)	Wash fastness		
	Assessment of change in color	Assessment of staining	
		Silk	Cotton
Undyed fabric	4	4-5	4-5
40	4	4-5	4-5
80	4	4-5	4-5
120	4-5	5	4-5
160	4-5	5	5
200	4-5	5	5
240	5	5	5

4. CONCLUSIONS

The present study was planned, to be looked out for a safer alternative for dyeing with natural dyes. Leaves of neem can be used as a dye for coloring textiles. It was found that neem dye can be successfully used for dyeing of silk fabric to obtain a wide range for soft hell shades. The process of extraction was simple and environmentally friendly. The natural colorant obtained from neem leaves has been successfully used as an eco-friendly dye to obtain different shades of yellow.

It was concluded that the absorption coefficient, extinction coefficient, color strength, tristimulus values and color parameters values were found to be influenced by either the concentration of the neem dyeing bath and exposure to artificial daylight for 160 h concerning the chemical structure of the used dye. Moreover, the dye-ability of silk fabrics was investigated. The increase in the dye-ability may be due to: the enhanced desorption of the dye, the greater availability of the dye molecules in the vicinity of the fiber, and the increase in diffusion due to the increase in the amorphousity of the fiber. Broad variations in shade and color depth were achieved, which indicate that the dye is environmentally and ecological acceptable for dyeing technology. Moreover, neem plant shows good shad reproducibility with significant fastness properties.

It was mentioned that, it is the first time that Egyptian silk fabrics were dyed with neem dyed extract. Also, it is the first time an optical technique investigated this search.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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