Optimization of Dyeing of Wool with Madder and Liposomes by Central Composite Design

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ABSTRACT: The preparation and production of multilamellar liposomes from Soya lecithin with 75% phosphatidylcholine were carried out and the behavior of liposomes in dye-bath at different temperature, time, and concentration were examined. It was found that liposomes with concentration of below 3% o.w.f. (on weight of fabric) in the dye-bath increases the amount of K/S for the samples dyed at 85°C or below 85°C for 60 min. Dyeing of wool at higher temperature and longer time with higher concentration of liposomes reduces the color strength. The results showed that using 2% o.w.f. of liposomes in dye-bath at 85°C for 60 min improves the K/S. The Central Composite Design is used for the experimental plan with three variables on the results of color strength. Statistical analysis confirms the optimum conditions obtained by the experimental results. It was also found that wash, light, wet and dry rub fastness properties of samples dyed with madder including liposomes have not changed significantly. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 106: 1614–1621, 2007

Key words: liposomes; wool dyeing; color strength (K/S); fastness

INTRODUCTION

Low temperature of wool dyeing has benefits such as lower energy conservation and wool fibers protection by either decreasing the temperature or shortening the processing time at high temperature during dyeing.[1] The wool fabric dye at low temperature has both more natural feeling and improved durability, using some of the known synthetic auxiliaries in dye-bath during low-temperature dyeing.[1]

Liposomes are spherical synthetic layers of phospholipids, which has been formed like closed vesicles with an aqueous core and ranging from 10 nm to 10 μm in diameter.[2,3] Liposomes compose of lipid vesicle bilayers enclosing a volume. These structures have hydrophobic and hydrophilic parts. The hydrophilic part is composed of phosphate and choline groups, and the hydrophobic part is made up of hydrocarbon chain.[4] Phosphatidylcholine is the most widely used in biological lipid for producing liposomes.[1]

Wool dyeing and wool blends with liposomes have demonstrated to improve quality, energy conservation, and lower environmental impacts.

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Recently, commercial liposomes were incorporated into textile auxiliaries, mainly for wool dyeing.[5–7] This is a clean technology that has already been adapted by some textile industries. These are additional benefits for material-weight yield during subsequent spinning. These improved smoothness and mechanical properties of the dyed textiles, and showed a clear reduction in the contamination load of the dye-baths.[8]

Use of liposomes as an auxiliary in wool dyeing can be related to the bilayer structure of lipids from the cell membrane complex (CMC) of wool that is similar to the liposomes and the action of this morphological fraction of the fiber in wool processing.[4] A wool fiber includes of cuticle and cortical cells held together by the CMC and forms the continuous phase in the keratin.[9] This phase contains a small amount of lipid material. Diffusion properties of wool fibers are influenced by the lipid structure of the intercellular spaces that could act as ''solvents'' for hydrophobic chemical. The dyes diffuse with ease into swollen regions such as the CMC (intercellular diffusion) rather than through the cuticle cells (transcellular diffusion).[10]

Last few years, several articles have related the potential application of liposomes in wool dyeing. Meza et al. have investigated liposomes as doer in wool dyeing with acid,[11,12] disperse[13,14] and metal complex dyes.[15] Also they have worked on the

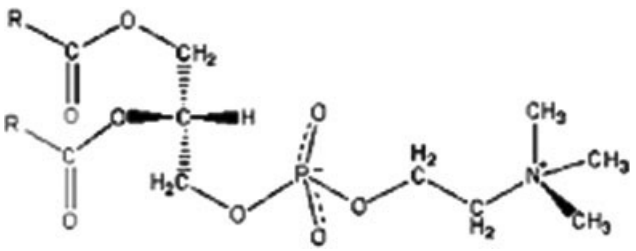

Figure 1 Chemical structure of the phosphatidylcholine.

effects of commercially available liposomes as a simple additive.[1,15,16] Recently they used an optimized mixture of commercial liposomes and cationic surfactant to improve leveling property.[17] In the previous article, the influence of temperature on stability of multilamellar liposomes (MLV) in wool dyeing was studied, and it was found that the presence of 1% o.w.f. (on weight of fabric) of liposomes at 85°C could improve the dye exhaustion of Irgalan Blue FBL on wool fabric. It has also reported that the wash fastness properties of dyed samples with liposomes have also improved.

There is no report on using liposomes in wool dyeing with natural dyes. Therefore, we try to prepare and produce MLV from Soya lecithin with 75% phosphatidylcholine and study the influence of liposomes in dye-bath at different temperature, time, and concentration during wool dyeing with madder as a most famous natural dye. The dyeing temperature and time were optimized with optimum concentration of liposomes, and the morphology of the liposomes dyed samples has been investigated by scanning electron microscope (SEM). The wash, light, wet and dry rub fastness properties of samples have also been reported.

MATERIALS AND METHODS

The wool fabric with plain woven structure from 48/2 Nm yarns was supplied by Iran Merino. The fabric was scoured with 1% anionic detergent VEROLAN-NBO (supplied by Rodulf) at 70°C for 45 min, and then washed with tap water, and dried at room temperature. Industrial grade of aluminium sulphate was used for mordanting of wool samples. Soya lecithin (containing 75% phosphatidylcholine) with phase transition temperature (T_c) of −18°C was gifted by Lipoid (Germany). The chemical structure of the phosphatidylcholine is illustrated in Figure 1.

Madder was prepared from Yazd providence of Iran. The chemical structure of important dyes in madder is illustrated in Figure 2.

The reflectance spectra of the dyed samples were recorded on an ACS Spectra Sensor II integrated with an IBM-PC. The wash-fastness of the liposomes treated madder-dyed fabric were measured according to ISO 150-C01. For light-fastness measurements, the samples were exposed to the daylight for 7 days according to the daylight ISO 105-B01, and changes in the color (fading) were assessed by the blue scale. Also the dry and wet rub fastness of the samples evaluated according to ISO 105-X12. The sample pictures were taken with Philips XL30 SEM with ×4000. The drop absorbency of the fabric samples was also measured by dropping of water droplet from 1 cm on the fabric surface on the glass by a small syringe. The time of complete absorption of the water droplets on the fabric surface was recorded and the mean value of 20 replicates was reported.

Liposomes preparation

MLV liposomes were prepared following the thin-film hydration method. A lipid film was formed by

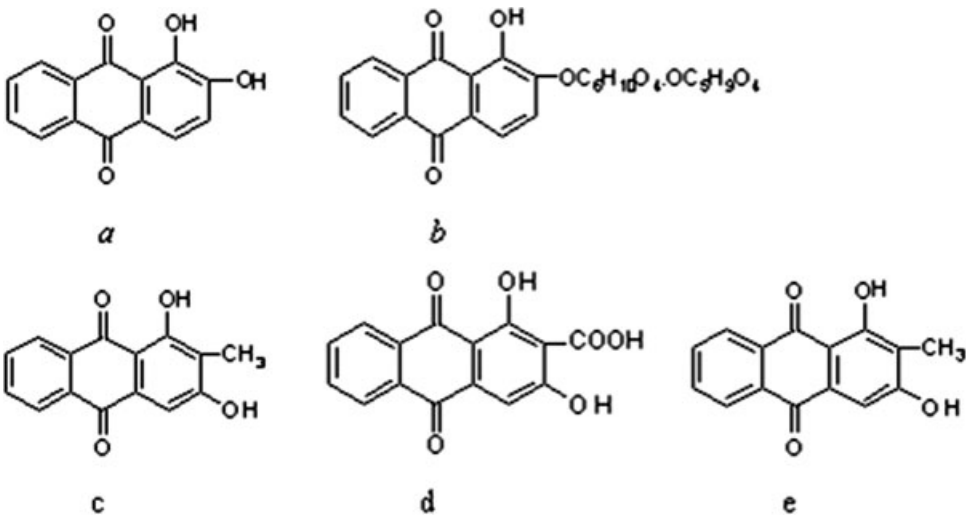

Figure 2 Chemical structure of important dyes in madder: (a) alizarin, (b) Rubierythric acid, (c) Rubiadin, (d) Munjistin or Purpuroxanthincarbloxylic acid, (e) Purpuroxanthin or Xanthopurpurin.[17]

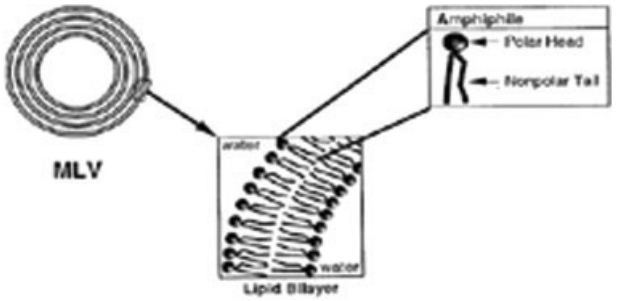

Figure 3 Schematic structure of MLV liposomes.

removing the organic solvent with rotary evaporation (with temperature of bath being 35–40°C and 30 rpm) from a chloroform solution containing Soya lecithin. Aqueous phase containing distilled water was added to the lipid film. The solution was shaken by hand to deliver the lipid from the walls of flask and disperse large lipid aggregates; glass beads were added to facilate dispersions. The milky suspension was agitated at 40°C to obtain a complete emulsion. This means that the lipid extensively hydrated and MLV liposomes formed.[1] The schematic structure of liposomes is illustrated in Figure 3.

The aggregation states of the vesicles were estimated as a measure of the physical stability of the liposomes suspensions. This was done by monitoring the variations of vesicle size in different temperature (40, 50, 60, 75, 85, and 95°C). An optical microscope was used to show the changes of liposomes in lipid phase as function of temperature.

Preparation for dyeing

Before dyeing, the wool samples should be cleaned to prepare the samples free from the impurities. Therefore the samples are scoured in first step and then dyed later. Also the dyestuff should be ready for process too. We can extract dyestuff from the natural collected madder.

Scouring

The samples were scoured in a bath containing 1 g/L anionic detergent, 1 ml/L ammonia (pH = 8.5) in 70°C for 45 min with liquor-to-goods ratio (L : G) of 40 : 1. The samples were then rinsed with warm water (60°C) and tap water and then dried at room temperature.

TABLE I
Ranges of Variables

Variable	Lower limit	Upper limit
Temperature (°C)	75	95
Time (min)	30	60
Liposome (%)	0	3

TABLE II
Central Composite Design for Dyeing of Wool with Madder

Run number	*A*: Temperature (°C)	*B*: Time (min)	*C*: Concentration (mg/mL)	*Y*: Color, *K*/*S*
1	75.00	30.00	1.00	8.31
2	90.00	30.00	1.00	23.96
3	75.00	60.00	1.00	14.84
4	90.00	60.00	1.00	23.94
5	75.00	30.00	3.00	9.27
6	90.00	30.00	3.00	22.91
7	75.00	60.00	3.00	17.81
8	90.00	60.00	3.00	22.65
9	69.89	45.00	2.00	9.32
10	95.11	45.00	2.00	22.56
11	82.50	20.00	2.00	18.43
12	82.50	70.00	2.00	24.61
13	82.50	45.00	0.32	18.62
14	82.50	45.00	3.68	20.9
15	82.50	45.00	2.00	20.6
16	82.50	45.00	2.00	20.37
17	82.50	45.00	2.00	20.94
18	82.50	45.00	2.00	20.84
19	82.50	45.00	2.00	20.7
20	82.50	45.00	2.00	20.79

Dyestuff extraction

For extraction of dyestuff, the madder were steeped in water solution for 24 h and then heated at 70°C for 20 min, and the solution was then passed through the filter. The filtered solution was transferred to a glassing flask. The solution of dye was concentrated by removing the water with rotary evaporation. The glassing flask was weighed before and after experiment and the weight of extracted dye obtained by the following formula:

$$W_{\text{ext.}} = W_a - W_b$$

$W_{\text{ext.}}$ is the weight of extracted dye (g) and W_a and W_b are the weight of flask after and before processing, respectively.

TABLE III
Regression Coefficients and Determination Coefficient

Y coefficient	Color strength, *K*/*S*
b_0	−348.8893
b_1	7.18647
b_2	1.47100
b_3	10.54856
c_1	−0.033761
c_2	3.27948 E − 002
c_3	−0.54847
b_{12}	0.017056
b_{13}	−0.10450
b_{23}	0.014750
R^2	0.9741

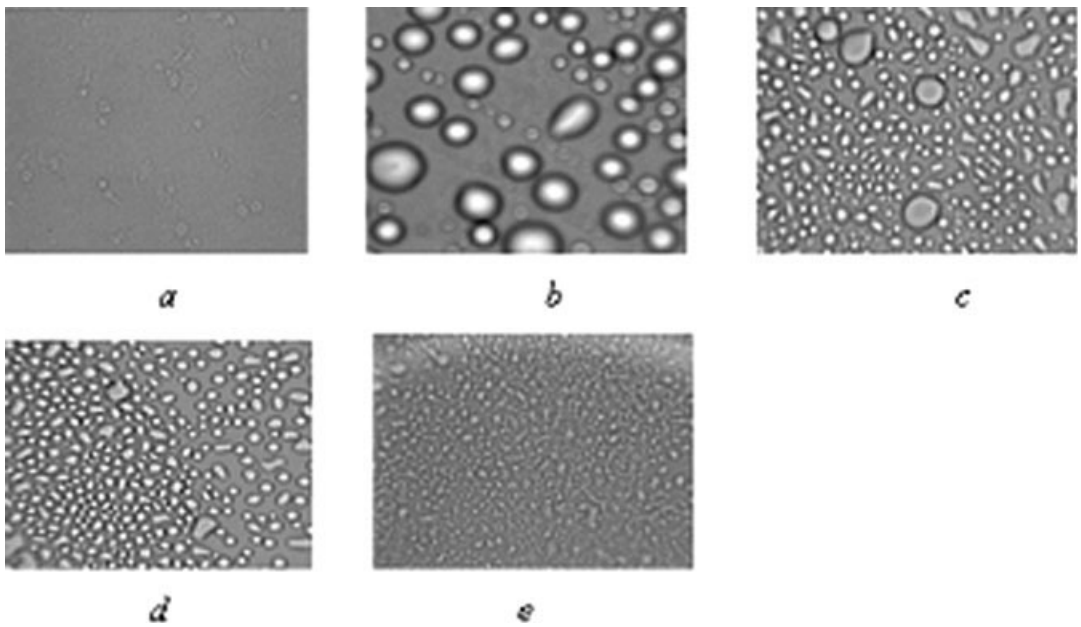

Figure 4 Liposomes solution with 20 mg/mL concentration at (a) 50°C, (b) 60°C, (c) 75°C, (d) 85°C, and (e) 95°C (×100).

Mordanting

The scoured samples (L : G = 40 : 1) were steeped in the mordant bath prepared with 20% o.w.f. of aluminium sulphate with pH = 4.5–5.8 (adjusted by acetic acid).

Mordanting of sample was started at room temperature and the temperature was raised for 2°C/min to boil and heated for 60 min. The samples were rinsed with tap water and dried at room temperature.

Dyeing

The mordanted wool samples were steeped in the dye bath with liquor-to-goods ratio of 40 : 1 that was prepared by 2% o.w.f. of extracted dye at pH 4.5–5.5 (acetic acid) with different concentrations of freshly prepared MLV liposomes (0, 1, 2, 3% o.w.f.).

Dyeing was started at room temperature and then raised 2°C/min to the final desired temperature including 75, 85, and 95°C. The dyeing was carried out with liposomes and without liposomes in various times of 30, 45, and 60 min. The samples were rinsed with tap water and dried at room temperature.

The amount of reflectance was selected at the maximum wavelength and the K/S value was calculated according to the Kubelka–Munk equation:

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

Experimental design

The Central Composite Design used for experimental plan with three variables is shown in Table I. Three variables including liposomes amount, time, and temperature were studied. The ranges of these variables are shown in Table I. Details of Central Composite Design for dyeing of wool with madder are demonstrated in Table II. Also the influence of the variable on the results Y [color strength (K/S)] is adjusted using the following second order polynominal function:

$$Y = b_0 + \sum b_i X_i + \sum b_{ij} X_i X_j + \sum c_i X_i^2 \quad i \geq j \qquad i, j = 1, 2, 3$$

In this equation, b_0 is an independent term according to the mean value of the experimental plan, b_i are regression coefficients that explain the influence of the variables in their linear form, b_{ij} are regression coefficients of the interaction terms between variables, and c_i are the coefficients of quadratic form of variables. Equation regression coefficients b_i, b_{ij}, c_i and the determination coefficient R^2 are shown in Table III.

Therefore, the final model is:

$$\begin{aligned} K/S = {} & -348.88931 + 7.18647^*A + 1.47100^*B \\ & + 10.54856^*C - 0.033761^*A^2 + 3.27948\ \mathrm{E} \\ & - 002^*B^2 - 0.54847^*C^2 - 0.017056^*A^*B \\ & - 0.10450^*A^*C + 0.014750^*B^*C \end{aligned}$$

In this equation, A, B, and C are temperature (°C), time (min), and concentration (mg/mL), respectively.

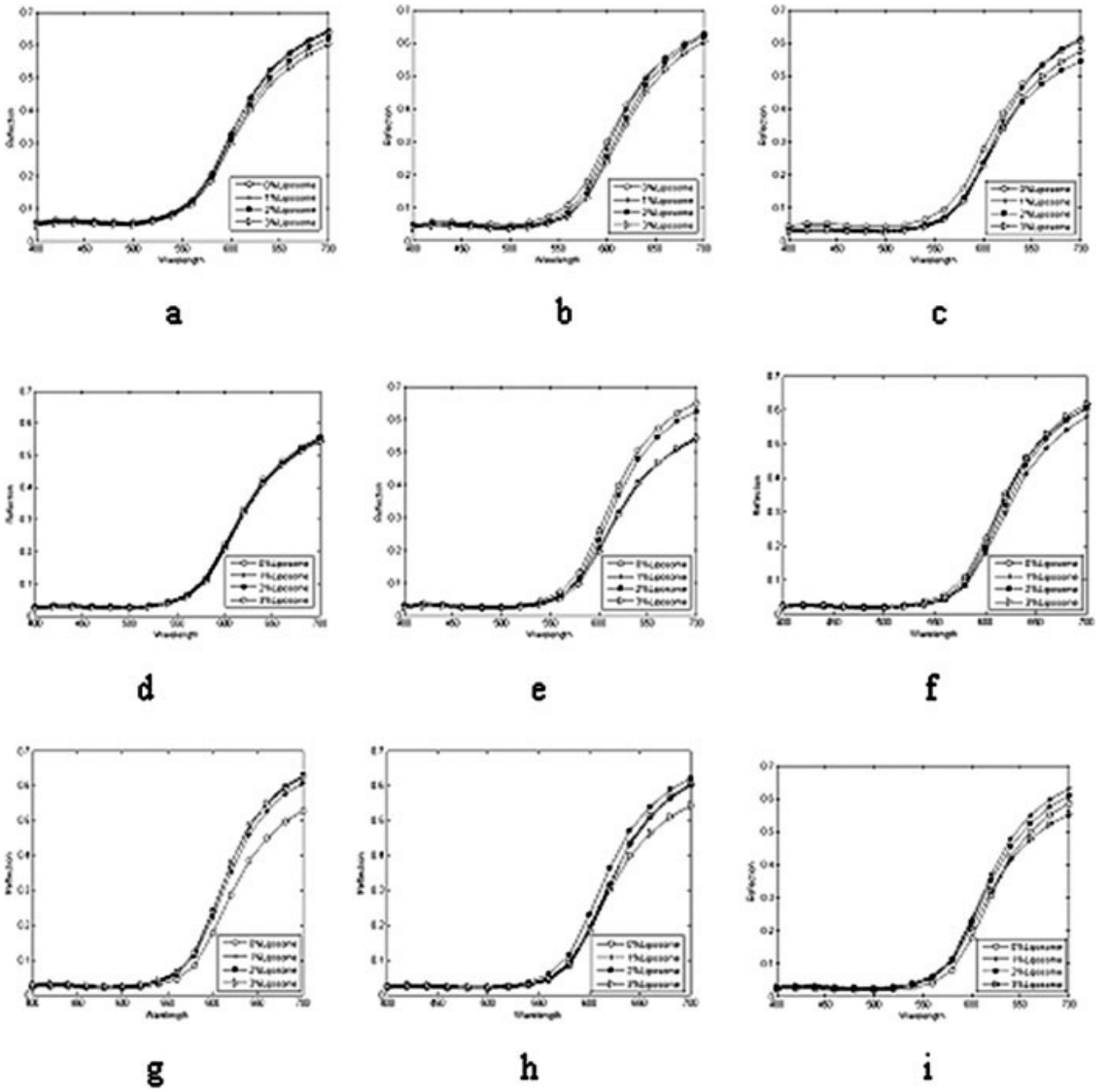

Figure 5 The reflectance spectrum of the madder-dyed sample at (a) 75°C for 30 min, (b) 75°C for 45 min, (c) 75°C for 60 min, (d) 85°C for 30 min, (e) 85°C for 45 min, (f) 85°C for 60 min, (g) 95°C for 30 min, (h) 95°C for 45 min, (i) 95°C for 60 min with different concentration of liposomes.

RESULTS AND DISCUSSIONS

Stability of liposomes

To determine the stability of prepared liposomes, verity of liposomes size due to aggregation or solubilization was monitored by measuring the variations in mean vesicle size distribution during preparation of liposomes.

The results of micrograph for 20 mg/mL liposomes showed that preparation of liposomes at 50°C can be done without any changes in the particles size of liposomes distribution due to the aggregation [Fig. 4(a)].

Increasing of temperature above 50°C converted the liposomes into large particles of phospholipids at 60–75°C [Fig. 4(b,c)], and above 75°C the large particle of phospholipids transfer to smaller particles [Fig. 4(d,e)]. These changes of the particles size of liposome leads to producing of a uniform layer of phospholipids on the wool fabric surface above

TABLE IV
***K/S* Values of the Dyed Samples with Different Liposomes Concentration and Time and at 75°C**

Liposomes concentration (%)	*K/S*		
	30[a]	45	60
0	7.89	9.28	10.70
1	8.31	9.55	10.53
2	8.20	11.71	17.25
3	9.27	12.63	17.81

[a] Numbers 30, 45, and 60 represent time (min).

TABLE V
***K/S* Values of the Dyed Samples with Different Liposomes Concentration and Time at 85°C**

Liposomes concentration (%)	*K/S*		
	30[a]	45	60
0	17.87	18.62	23.11
1	17.89	19.92	24.66
2	17.80	20.94	25.82
3	20.72	20.97	24.45

[a] Numbers 30, 45, and 60 represent time (min).

TABLE VI
***K/S* Values of the Dyed Samples with Different Liposomes Concentration and Time at 95°C**

Liposomes concentration (%)	*K/S*		
	30[a]	45	60
0	22.93	25.76	25.96
1	21.54	25.05	21.59
2	21.07	22.56	19.74
3	20.95	2.38	19.03

[a] Numbers 30, 45, and 60 represent time (min).

85°C. This also leads to a decrease in the amount of *K/S*. From these results, it can be concluded that the most ability of liposomes as doer in dyeing can be obtained at the 40–80°C.

Dyeing profiles

The color strengths (*K/S*) were measured by the spectrophotometer (Texflash from Data color). The reflection of the samples was measured at 400–700 nm wavelengths. All of the madder dyed samples indicated the low value of reflectance at 485 nm (λ_{max}) (Fig. 5). It can be seen in Figure 5 that the reflectances of different samples are similar and they indicated the same curves.

The *K/S* values were obtained on the samples applied with liposomes concentration from 0 to 3% o.w.f. and madder dye on the aluminum mordanted wool at three different temperatures (75, 85, and 95°C) and time (30, 45, and 60 min). It can be observed from Tables IV and V that any increase in the time of dyeing and concentration of liposomes

TABLE VII
Wash Fastness of Dyed Samples

Sample cotton	Wash fastness	Staining on wool	Staining
Dyed without liposome at 95°C for 60 min	4–5	5	5
Dyed with liposome at 85°C for 60 min	4	5	5

TABLE VIII
Dry and Wet Rub Fastness of the Dyed Samples

Sample	Dry rub fastness	Wet rub fastness
Dyed without liposome at 95°C for 60 min	3–4	4–5
Dyed with liposome at 85°C for 60 min	3–4	4–5

TABLE IX
Light Fastness of the Dyed Samples

Sample	Light fastness
Dyed without liposome at 95°C for 60 min	4–5
Dyed with liposome at 85°C for 60 min	4–5

caused an increase in the values of *K/S*. The results indicate that the temperature is more effective than both time of dyeing and liposome concentration at 75 and 85°C. The values of (*K/S*) continue to increase at 85°C for 60 min with 2% o.w.f. liposomes concentration. It seems that liposomes apply as doer for dye at these conditions.

Increasing of temperature to 95°C with different time and liposomes concentrations decreases the value of *K/S* (Table VI). This could be related to the liposomes stability. The liposome at 95°C converted to the smaller particles of phospholipids. These changes of the particle size of phospholipid lead to coating of wool surface with a layer of phospholipid above 85°C, which leads to decrease the value of *K/S*. The results also indicated that the samples dyed without liposomes have a higher value of *K/S* comparing with the sample dyed with liposomes above 85°C. According to Table V, it may conclude the same results as color strength results. It means

TABLE X
ANOVA for Response Surface Quadratic Model

Source	Sum of squares	DF	Mean square	*F* value	*P* > *F*
Model	452.60	9	50.29	41.87	<0.0001
A	314.12	1	314.12	261.52	<0.0001
B	46.44	1	46.44	38.66	<0.0001
C	2.15	1	2.15	1.79	<0.04
A^2	51.97	1	51.97	43.27	<0.0001
B^2	0.078	1	0.078	0.065	<0.034
C^2	4.34	1	4.34	3.61	<0.028
AB	29.45	1	29.45	24.52	<0.0006
AC	4.91	1	4.91	4.09	<0.045
BC	0.39	1	0.39	0.33	<0.01
Residual	12.01	10	1.20		
Lack of fit	11.81	5	2.36	56.90	
Pure error	0.20	5	0.41		
Cor. total	464.61	19			

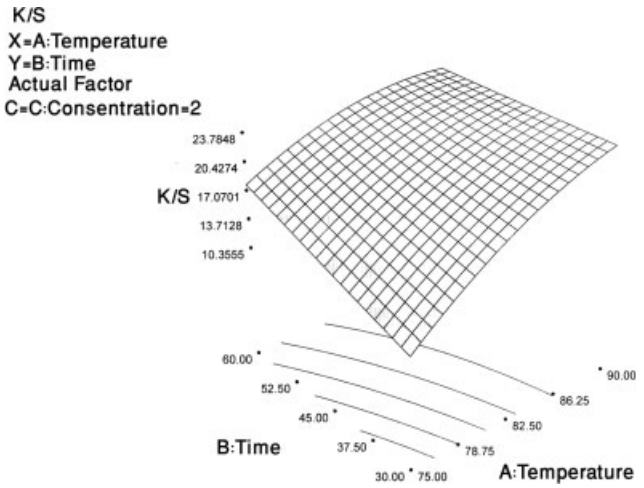

Figure 6 Design of expert plot.

that higher reflectance leads to lower absorption and lower color strength.

It can be concluded that utilization of 2% o.w.f. liposomes in dye-bath at 85°C for 60 min, reduces the temperature of dyeing about 10°C compared with a conventional dyeing process. These results are in accord with the results obtained by de la Maza et al. for the dyeing of wool with metal complex dyes.

To study the effect of liposomes on the dyed wool samples, wash, light, wet and dry rub fastness were tested and the results listed in Tables VII–IX. The results revealed that the wash, light, and rub fastness of the samples dyed along with liposomes are similar to the samples dyed without liposomes. However, the wash fastness of the samples dyed along with liposomes improved marginally, which can be due to the lipid precipitation on the fabric surfaces. This can be acted as a barrier against bleeding of the dye from the fabric.

Statistical analysis

The analysis of variance (ANOVA) is given in Table X. It can be concluded that all of the terms in this model are significant. Also the lack of fit test with the *P*-value of 0.075 shows the model is significant and it is fitted well.

According to the ANOVA results, the fitted model is:

$$K/S = -348.88931 + 7.18647^{*}A + 1.47100^{*}B + 10.54856^{*}C - 0.033761^{*}A^2 + 3.27948\ E - 002^{*}B^2 - 0.54847^{*}C^2 - 0.017056^{*}A^{*}B - 0.10450^{*}A^{*}C + 0.014750^{*}B^{*}C$$

Figure 6 also shows the response surface of the model. By using Design Of Expert software the optimum design point with desirability of 94.60% is about temperature of 85°C, time of 60 min, and liposome concentration of about 2%.

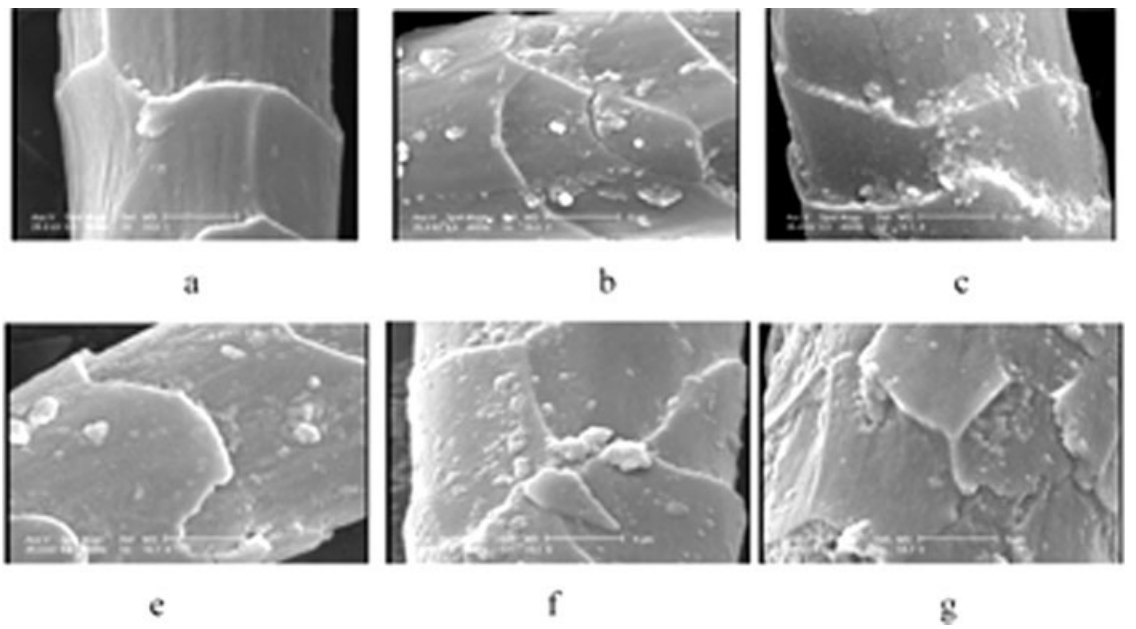

Figure 7 SEMs of (a) raw wool, (b) Aluminum mordanted, (c) treated with 2% liposome, (d) Al. mordanted dyed with madder for 60 min at 95°C, (e) Al. mordanted dyed with madder and 2% liposome for 60 min at 85°C for 60 min, (f) same as (e) with 4% liposome (×4000).

TABLE XI
The Mean Values of Water Drop Absorption Time on the Different Wool Fabric Samples

Sample	Mean of drop absorption time (s)
Raw wool	294
Dyed with liposome at 85°C for 60 min	540
Dyed without liposomes at 95°C for 60 min	486

SEM

SEM was utilized in considering of liposomes effect and mordant on the wool fabric surfaces. The results in Figure 7 show a normal morphological form for the raw wool and scales clearly has been seen [Fig. 7(a)]. For the mordanted wool some particles of mordant has been observed on the fiber surface [Fig. 7(b)]. The wool sample treated with 2% o.w.f. of liposomes indicated an aggregation of phospholipids on the edge of scales [Fig. 7(c)]. The surfaces of the wool sample dyed with optimum conditions (85°C, 60 min, and 2% o.w.f. liposomes) indicated again some particles on the surface. These could be presumably produced by the mordant and liposomes that are scattering on the fiber surface randomly. The picture of the sample dyed without liposome indicated the same particles on the fiber surface, but the coating on the sample dyed with liposomes is more than without liposomes. Also the liposomes remaining on the edge of the scale of the fiber dyed with liposome has been clearly observed.

Water drop absorption test

The results indicated that the time required for raw wool sample was 294 s, for mordanted and dyed wool was 486 s, and for liposome dyed 540 s (Table XI).

The results show that mordanting and dyeing process on wool reduce the water absorption and the wool fabric becomes more hydrophobic. However, the hydrophobicity is higher for the samples dyed along with liposomes. This is because of coating of the fibers surface by the phospholipids of the collapsed liposomes.

CONCLUSIONS

Liposomes are environmental friendly compounds that can be used in dyeing of wool. Applications of liposomes as an assistant or carrier in dyeing of wool by some of the synthetic dyes have already been reported. However, the dyeing of wool dye with natural dyes is an interesting and new subject. Liposomes in dyeing of wool with madder show a clear reduction in dyeing temperature. The mechanism action of liposomes in wool dyeing with madder is similar to the dyeing of wool with synthetic dyes, and it has the same influence on the dyeing process. It can be hypothesized that the liposomes apply as a carrier of dye and are absorbed by the wool fiber, and the interaction between the lipid concentration of liposomes and CMC of wool takes place. This leads to accelerate dye diffusion and dye uptake at 85°C. However, above this temperature the liposomes collapsed and covered the wool fibers. Thus acting as a barrier for dye uptake and resulting in reduced K/S. Statistical analysis by DOE indicated that application of 2% liposome at 85°C for 60 min on wool dyeing with madder produces an optimum design point with desirability of 94.60%.

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