

## Optimizing a wool dyeing process with reactive dye by liposome microencapsulation

M.M. El-Zawahry <sup>a,\*</sup>, S. El-Shami <sup>b</sup>, M. Hassan El-Mallah <sup>b</sup>

<sup>a</sup> Textile Research Division, National Research Centre, El-Behouth St. Dokki, Cairo, PO 12622, Egypt

<sup>b</sup> Food Technology and Nutrition Division, National Research Centre, El-Behouth St. Dokki, Cairo, PO 12622, Egypt

Received 7 September 2005; received in revised form 16 December 2005; accepted 24 April 2006

Available online 11 July 2006

### Abstract

Phospholipids mixture from commercial soybean lecithin (SBL) was prepared as acetone insoluble fraction (AIF) which was acetylated at 70%. Phospholipids composition was elucidated with TLC-Densitometry. The acetylated AIF was used for preparing liposomes for encapsulating reactive dye for wool and wool blends dyeing. Ultrasonic irradiation was used in preparing liposomes. Particle size measurements of the liposomes encapsulating the dye at different concentrations were measured. Maximum particle size of about 16 nm was obtained at liposome concentration of 4 g/l.

Dyeing was conducted under different parameters in presence and absence of microencapsulated (MS) dye. The colour strength values, exhaustion, levelling and fixation percent were higher in presence of liposome and tensoactive product than those of conventional dyeing process. These more satisfactory results of wool dyeing encouraged the application of the optimum conditions of wool dyeing on wool blend fabrics with efficiency dye uptake and levelling. Moreover, microencapsulation with liposomes as a new efficient technology for dyeing process prevents environmental pollution.

© 2006 Elsevier Ltd. All rights reserved.

**Keywords:** Soybean lecithin; Liposome vesicles; Nanoemulsion; Dyeing; Wool; Reactive dye

### 1. Introduction

Encapsulation or liposome technology is applied in numerous fields, such as in pharmaceuticals, cosmetics, foods, detergents, textiles and other applications where it is important to liberate the encapsulated material slowly. Liposomes can be defined as any structure composed of lipid vesicle bilayers that enclose a volume and liposomes containing a dye are generally large, irregular, and unilamellar. From a chemical point of view, the liposome or phospholipid vesicle is an amphoteric compound containing both positive and negative charges. Therefore, the use of liposomes has been examined for delivering dyes to textiles in an environmentally sensitive way leading to technologically useful results [2–6]. In textiles, the major

interest in microencapsulation is currently in the application of durable fragrances and skin softeners. Other applications include insect repellants, dyes, vitamins, antimicrobial agents, phase-change materials and medical applications, such as antibiotics, hormones and other drugs. The potential applications of microencapsulation in textiles are as wide as the imagination of textile designers and manufacturers [1]. On the other side, there is an increasing interest in the textile industry in ecofriendly textile processing, in which the use of naturally occurring materials such as phospholipids becomes very important. In recent years liposomes from phospholipids as a biological material have been widely used in dyeing process. In the previous work it was reported that PCs are the most prevalent among the various phospholipids for the preparation of stable liposomes as a dye carrier, dispersing agent and levelling agent in dyeing process [3,4,7–9]. Early studies showed that phosphatidylcholines (PCs) are obtained from different sources such as egg yolk and SBL mixed with other phospholipids. Also

\* Corresponding author. Tel.: +20 23371718; fax: +20 23370931.

E-mail addresses: [zawahry\\_m@hotmail.com](mailto:zawahry_m@hotmail.com), [manalzawahry@yahoo.co.uk](mailto:manalzawahry@yahoo.co.uk) (M.M. El-Zawahry).

hydrogenated or synthesized PCs have been more commonly used due to their high chemical stability [10].

Meanwhile, few studies were reported on using PC liposomes for dyeing wool with reactive dyes, a dye type with great potential, once the problems of levelling and fixation are overcome. There has not been any work reported on using liposomes for dyeing wool/blend fabrics with reactive dye in one-bath technique.

The main objective of this work is to prepare phospholipids of certain composition to be acetylated to certain degree for forming liposomes capable of encapsulating reactive dyes (LERD) for wool and wool/blend fabrics dyeing. The technology of microencapsulation is selected since it has given rise to a number of innovations in textile domain as not cost effective and simple dyeing process that has lower impact on the environment. It was feasible to achieve liposome characteristics regarding holding the reactive dyes in stable conditions so that their release, as occasion demands, are fulfilled in the first stage of dyeing process. Phospholipids mixture, containing principally phosphatidylcholine (PC) and phosphatidylethanolamine (PE) was prepared from commercial SBL as AIF that subsequently modified via acetylation under controlled reaction conditions. This can help obtaining nanoparticle liposomes with possibly higher quantity of the encapsulated reactive dye.

Properties of wool dyeing with nanoencapsulated reactive dye, such as *K/S* values, exhaustion percent, levelling effect, reduction in dye auxiliaries and fixation parameters of the encapsulated reactive dyes will be evaluated. Heterobifunctional Sumifix Supra Brilliant Red 2BF reactive dye is selected for dyeing wool and wool/blend fabrics. The properties exhibited by the conventional dyeing and liposome encapsulated reactive dye were compared.

## 2. Experimental

### 2.1. Materials

#### 2.1.1. Fabrics

Mill scoured 100% wool fabric of 500 g/m<sup>2</sup> supplied by Misr Co. for Spinning and Weaving (Mehalla El-Kubra) was used. Before dyeing, the fabric was treated in aqueous solution containing 2 g/l sodium carbonate and 5 g/l non-ionic detergent (Hostapal CV, from Clariant – Egypt) for 30 min at 50 °C, then washed thoroughly with water and dried at room temperature.

Mill scoured wool blended fabrics e.g. polyester/wool (55:45) of 20 g/m<sup>2</sup> were obtained from Misr Mehalla Co., acrylic/wool (67:33) of 20 g/m<sup>2</sup> obtained from Wooltex Co. – Egypt and cotton/wool (70:33) of 20 g/m<sup>2</sup> blend fabric was delivered from Golden Tex Co., Tenth of Ramadan – Egypt. These blend fabrics were washed or treated in aqueous solution containing 2 g/l non-ionic detergent and 0.5 g/l sodium carbonate at 60 °C for 45 min. Finally they were rinsed thoroughly and air dried at room temperature.

#### 2.1.2. Liposome formation

Commercially available SBL, supplied by Extracted Oils and Derivatives Company, Alexandria— Egypt, was used.

### 2.1.3. Dyestuffs and chemicals

Sumifix supra Brilliant Red 2BF (C.I. Reactive Red 194) was supplied by Sumitomo Chemical Co. and Triton X-100 (*t*-Octylphenoxy polyethoxyethanol) was obtained from Sigma Aldrich. All other chemicals used in this study were of laboratory reagent grade.

## 2.2. Methods

### 2.2.1. Preparation of AIF from commercial SBL

Crude soybean commercial lecithin was deoiled with acetone to prepare AIF enriched with phospholipids mixture. Thus 65 g portion of crude lecithin was agitated moderately with 130 ml cold acetone followed by two extraction steps using 70 ml acetone in each. The acetone layer containing neutral lipids was decanted and the AIF was dried under reduced pressure. The AIF contained mainly phospholipids and glycolipids whereas the acetone solution contained triglycerides, sterols, sterol esters tocopherols, carotenoids, monogalactoside diglycerides and sterylglucosides. The AIF was subjected to TLC analysis in comparison with the crude lecithin to ensure the presence of the main phospholipid components.

### 2.2.2. Chemical modification of AIF

Trials have been made to prepare chemically modified AIF, to obtain derivatives having higher emulsification properties to be tested for wool dyeing via liposome encapsulation process. These derivatives comprised hydroxylated, and acetylated AIF phospholipids. The preliminary selection of the derivative and its suitability for wool dyeing is based on the *K/S* values as will be referred in the text.

**2.2.2.1. Hydroxylation of AIF.** Hydroxylation of AIF (50 g), prepared from representative sample of different commercial SBL batches, was conducted using hydrogen peroxide (7.0 g, 30%) to react with the double bonds of the unsaturated acyls of the phospholipid molecules under the catalytic action of small amounts of lactic acid (1.5 g, 75%) at 50 °C while stirring for 2 h [11]. The reaction was monitored by iodine and *K/S* values determinations.

**2.2.2.2. Acetylation of AIF.** Chemical modification of AIF, prepared from representative sample of different commercial SBL batches, was carried out by acetylation [13]. Acetylation was conducted using different mole equivalents of acetic anhydride (from 1 to 2) on the basis of the PE content of the sample. The reaction was carried out at 50–55 °C for 2 h and the reaction mixture was dried in vacuum. The reaction was monitored by TLC to follow the degree of acetylation of PE ( $R_F = 0.43$ ) to form *N*-Acetyl PE ( $R_F = 0.65$ ). PE and *N*-Acetyl PE were quantified by TLC-Imaging Densitometry [12]. The conversion ( $\lambda$ ) was calculated from the decrease of PE and the increase of *N*-acetyl PE. The product obtained was used for the preparation of liposome vesicles capable of encapsulating the reactive dye in wool dyeing process.

### 2.2.3. Preparation of liposome vesicles with microencapsulated reactive dye

The dyebath was prepared first at room temperature containing 1% shade owf of C.I Reactive Red 194 with

liquor-to-goods ratio 50:1, various concentrations of sodium sulphate (0, 5, 10, 15, 20 and 30% owf) were used and pH of the dyebath was adjusted to 5 by diluted acetic acid. The prepared dye solution was then added to different concentrations of acetylated AIF (0, 0.25, 0.50, 1, 2, 4, 6, 8 and 10 g/l) to form the lipid suspension, and the resulting coloured suspension was sonicated (CREST benchtop 575 HT, ultrasonic cleaner bath with frequency 38.5 kHz) for 30 min at 50 °C to obtain an emulsion at different sonic power from 100 to 500 W.

#### 2.2.4. Dyeing conditions

Dyeing of wool fabrics with reactive dye emulsion and with conventional dyebath was carried out, without using any synthetic auxiliary products with liquor ratio 50:1 at pH 5, 10% sodium sulphate and the bath temperature raised to 100 °C over 40 min. After which time, dyeing at this temperature was continued for further 60 min; the samples were then cooled to room temperature and washed with an aqueous solution of detergent (5 g/l) at 50 °C for 20 min.

Dyeing wool/blend fabrics with heterobifunctional reactive dyes using one-bath technique in presence and absence of liposome encapsulated dye was done as mentioned above in dyeing wool fabrics. The samples were then cooled to room temperature and washed with an aqueous solution of detergent (2 g/l) at 50 °C for 20 min.

### 2.3. Measurements and analysis

#### 2.3.1. Chemical composition of phospholipids

Densitometry on chromatographic plates directly determines the concentration of the phospholipid compounds of the SBL and AIF analytes under the following conditions by Camag TLC-Scanner III (TLC-Imaging Densitometry):

- TLC plate 20 × 20 cm, Ready Made, E. Merck.
- Absorption: 500 nm
- Application mode: GAMAG Linomar IV.
- Developing solvent: Chloroform:Methanol:Water:Ammonium hydroxide (65:30:4:2 v/v).
- Detection of phospholipids: Iodine vapour.
- Scanning mode: TLC Scanner with Labdata System and CATS software with integrator.

Each phospholipid sample of the commercial SBL and AIF (10% solution) was applied in 5 different nanogram concentrations, as 5 tracks on one plate to obtain more reliable results. Scanning at 500 nm of the developed and iodine-visualized TLC plate was conducted, and the results of the 5 tracks – referring to the separated phospholipid components in AIF and SBL were separately recorded as mean value compositions. Recognition of the phospholipid components was done with the help of reference components run along side with the applied samples.

#### 2.3.2. Particle size measurement

Microencapsulated reactive dye emulsions with different concentrations of acetylated AIF (0–10 g/l) were freshly prepared. Therefore one drop of each emulsion was mounted on a copper grid covered by thin film of carbon, and after drying, the samples were examined by Electron Transition Microscope EM-10 (Model, Zeiss – West Germany) at high voltage (hV) 60 kV and with resolution  $\sim 10$  Å.

Measurement of the particle size diameter of the prepared microencapsulated reactive dye of each sample was measured using the Leica Qwin 500 Image Analyzer (LEICA Imaging Systems Ltd, Cambridge, England).

#### 2.3.3. Colour measurements

The relative colour strength ( $K/S$ ) of the dyed fabrics was measured by light reflectance technique using the Perkin–Elmer UV/vis spectrophotometer (Model, Lambda 3B) at maximum wavelength of the dye, 519 nm [14].

The percentage of dyebath exhaustion was measured on a Shimadzu UV/vis spectrophotometer at  $\lambda_{\max}$  of the dye used. The percentage of dyebath exhaustion (% $E$ ) was calculated according to the following equation:

$$\%E = \left[ \frac{A_0 - A_F}{A_0} \right] \times 100 \quad (1)$$

where  $A_0$  and  $A_F$  are the concentrations of the dyebath before and after dyeing, respectively.

#### 2.3.4. Measurement of dye fixation

The percentage of dye fixation (% $F$ ) was measured by refluxing the dyed samples in 1:1 DMF/water solution for 15 min to extract the unfixed dye. The colour strength ( $K/S$ ) values of the extracted and dyed samples were then measured spectrophotometrically at  $\lambda_{\max}$  and the extent of dye fixation ratio on wool was calculated according to Eq. (2) [15,16].

$$\%F = [(K/S)_2 / (K/S)_1] \times 100 \quad (2)$$

where  $(K/S)_1$  and  $(K/S)_2$  are the colour strengths of the dyed samples before and after stripping, respectively.

From the dyebath exhaustion ( $E$ ) and dye fixation ratio ( $F$ ), the total dye fixation ( $T$ ), which is the percentage of dye chemically bound relative to the total amount of dye used, was calculated using Eq. (3).

$$\%T = (\%E \times \%F) / 100 \quad (3)$$

## 3. Results and discussion

### 3.1. Phospholipids

The phospholipids composition of each commercial SBL and AIF sample was expressed as mean value of five replicates and the results are recorded in Table 1 and Fig. 1. The total phospholipids in AIF and SBL amounted, respectively, to 88.8 and 51.0% of the weight of each sample. With reference

Table 1  
Phospholipids composition of crude commercial lecithin (SB) and AIF as determined by Camag TLC-Scanner II Imaging Densitometry

| Sample         | Total phospholipids (%) | Phospholipid components (wt%) |      |      |         |      |      |                                  | Non-phospholipid components <sup>b</sup> | Unknown components |
|----------------|-------------------------|-------------------------------|------|------|---------|------|------|----------------------------------|--|--------------------|
|                |                         | Lyso-PC                       | PA   | PI   | Lyso-PE | PC   | PE   | <i>N</i> -Acetyl PE <sup>a</sup> |  |                    |
| Commercial SBL | 51.8                    | 1.0                           | 5.9  | 10.0 | 0.5     | 19.4 | 14.0 | 1.0                              | 40.2                                     | 8.0                |
| AIF            | 88.76                   | 2.0                           | 10.0 | 12.0 | 1.7     | 37.0 | 22.0 | 4.0                              | 5.0                                      | 6.3                |

<sup>a</sup> Including free sterols.

<sup>b</sup> Mainly TGs + free and acetylated sterylglucosides.

to the phospholipid profile of AIF, it was found that it contained 2.0% lysophosphatidylcholine (Lyso-PC), 10.0% phosphatidic acid (PA), 12.0% phosphatidylinositol (PI), 1.7% lysophosphatidylethanolamine (Lyso-PE), 37.0% PC, 22.0% PE, 4.0% *N*-Acetyl PE, 5.0% non-phospholipid components and 6.3% unknown components (PC/PE = 1.68).

Referring to quantified phospholipids of commercial SBL, the results showed the presence of 1.0% Lyso-PC, 5.9% PA, 10.0% PI, 0.5% Lyso-PE, 19.4% PC, 14.0% PE, 1.0% *N*-Acetyl PE, 40.2% non-phospholipid components and 8.0% unknown components.

On the other side, the phospholipids of AIF were subjected to chemical modification, via hydroxylation and acetylation to try to improve its chemical and physical properties. However, only the hydroxylated product gave unsatisfactory results as indicated from the lower *K/S* values. Therefore it was advisable to use the acetylated AIF to prepare the liposomes. Different degrees of acetylation, namely 40, 70 and 75% were determined on the basis of PE conversion to *N*-Acetyl PE. The *K/S* values of the products were calculated to select the optimum degree of acetylation in wool dyeing. It was found that as the degree of acetylation increased from 40 to 70%, the *K/S* value also increased from 7.36 to 8.46, however, at 70–75%, there was a negligible increase in *K/S* values from 8.46 to 8.64%. It was found that the degree of acetylation of 70% was the optimum degree that gave higher colour strength value (*K/S* value). It seems possible that free hydroxyl groups in Lyso-phospholipids and PI were acetylated besides the formation of *N*-Acetyl PE.

Therefore, liposomes prepared from the acetylated AIF were effective to encapsulate the reactive dyes and release

them at appropriate dyeing conditions, as it will be mentioned later.

### 3.2. Effect of ultrasonic power

The effect of ultrasonic power for dispersing the liposomes and subsequent encapsulation of the dye was studied at different power levels (100–500 W). As shown in Fig. 2 the *K/S* values of the microencapsulated heterobifunctional reactive dye increased with increasing the power level up to 300 W, then levelling occurred from 300 to 500 W. The result of *K/S* values indicated that at 300 W microencapsules with a smaller size distribution were obtained by ultrasound irradiation at 38.5 kHz than by mechanical agitation.

### 3.3. Effect of the time period of ultrasound irradiation

The preparation of LERD emulsion using ultrasound irradiation at 300 W was carried out at different intervals of time. Fig. 3 shows that the rate of dyeing by LERD using ultrasound irradiation was higher than that exhibited by mechanical agitation method. These results show that ultrasound irradiation between 15 and 30 min was efficient to obtain microencapsules with smaller size distribution as well as a good microencapsulation yield of the dye. In addition, higher *K/S* values were obtained between 90 and 180 min at 1000 rpm by using mechanical agitation method.

### 3.4. Effect of liposome concentrations on the microencapsulation efficiency

The effect of liposome concentrations (0–10 g/l of acetylated AIF at 95 °C for 60 min) on the colour strength of

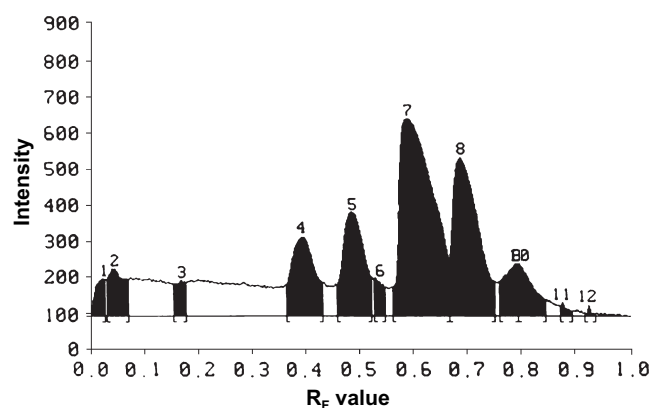


Fig. 1. Phospholipid composition of AIF by TLC-Imaging Densitometry at 500 nm. Peaks: 1 and 2, unknowns; 3, Lyso-PC; 4, PA; 5, PI; 6, Lyso-PE; 7, PC; 8, PE; 9 and 10, *N*-Acetyl PE and sterols; 11, SG; and 12, TG.

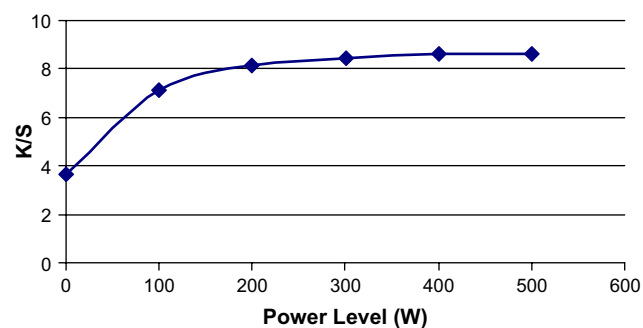


Fig. 2. Effect of ultrasonic irradiation power on the colour strength of dyed wool fabrics with microencapsulated C.I. Reactive Red 194.



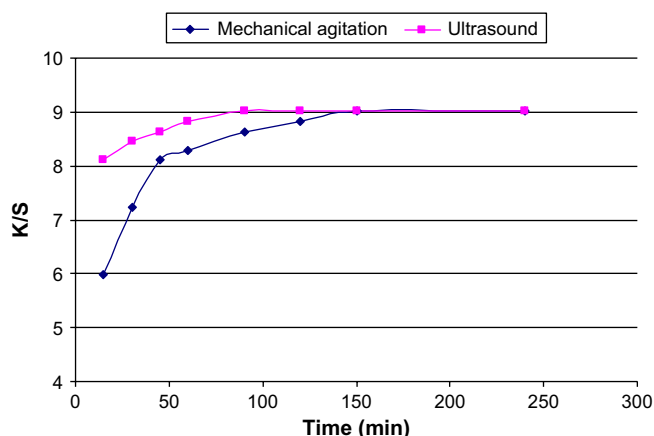


Fig. 3. Effect of agitation method on the colour strength of dyed wool fabrics with microencapsulated C.I. Reactive Red 194.

dyeing wool fabrics with microencapsulated (MS) heterobi-functional reactive dye (as an alternative to synthetic dye auxiliaries) were studied as shown in Fig. 5. Meanwhile, the tensoactive product Triton X-100 (20 g/l) was added after 30 min of the start of dyeing process [3] to destroy liposomes and to free the dye from it [17,18] and save energy and time.

It is worthy to mention that the average particle size of the commercial liposome was 50 nm, however, MS liposomes prepared in this work from acetylated AIF ranged from 7.59 to 16.56 nm as shown in Fig. 4. Fig. 5 indicates that both the  $K/S$  values and the exhaustion percent proportionally increased as the liposome concentration increased, reaching its maximum value at 4 g/l indicating higher encapsulation efficiency as it is clear from Fig. 6. The increase in  $K/S$  value may be due to the increase in the average particle size from 7.59 to 16.5 nm as the liposome concentration increased from 0.125 to 4 g/l. In other words the release rate of the dye from microcapsules increased when the latter were ruptured, and accordingly the dye could easily penetrate the wool fabrics and bound easily to the fibre. It is noteworthy to mention that increasing the concentration of liposomes above 4 g/l resulted in decreasing  $K/S$  values as well as exhaustion percent on wool fabrics throughout the dyeing process, i.e. retarding the rate of dyeing process. The results shown in Figs. 4 and 5 illustrate that the average particle size decrease monotonously from 12.33 to 9.24 nm as the concentration of liposomes increased from 6 to 10 g/l. However, as the concentration of

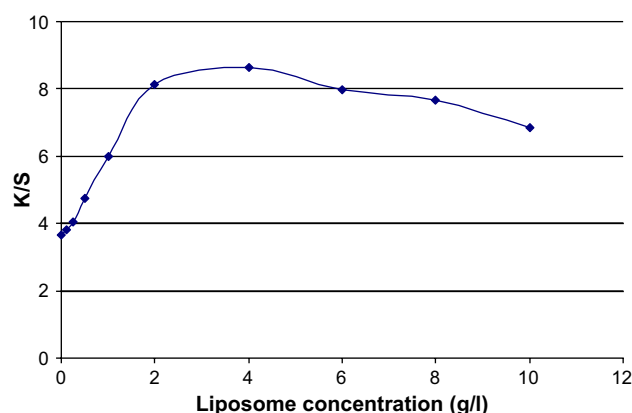


Fig. 5. Effect of various liposome concentrations on the colour strength of the dyed wool fabrics with microencapsulated C.I. Reactive Red 194.

liposome increased from 6 to 10 g/l the release rate of microencapsulated dye markedly decreased, which is accompanied by decrease in  $K/S$  values.

### 3.5. Levelling effect of liposome microcapsules

With references to the kinetic behaviour of dyeing wool fabric with LERD with different liposome concentrations on the levelling of the dye distribution, the present investigation showed that the use of liposomes of acetylated AIF resulted in inhibiting the dye exhaustion in the first stages of the dyeing process. Thus with the use of microencapsules' technique in all cases, the levelling effect of the dye was improved more than that obtained when using conventional auxiliaries. Meanwhile, the rate of exhaustion increased by adding 20 g/l Triton X-100, 30 min after the beginning of dyeing process. It seems that the dye was freed from the liposomes after 30 min of the dyeing time, at higher temperature (95 °C) and therefore redistribution of the dye between the fibres was enhanced, these results were in good accord with those reported by some authors [3,17,19]. Therefore, it can be suggested that the liposomes of acetylated AIF are the most suitable ones as levelling agent in industrial wool dyeing.

### 3.6. Stability of liposomes

Acetylated AIF possess higher emulsification properties and exhibit thermal stability [13]. To confirm the stability influence of acetylated liposomes on  $K/S$  values, dyeing was done at different reaction conditions (temperature, dyeing time and various liposome concentrations). The technique of adding Triton X-100 after 30 min of dyeing process was studied to destroy liposomes and hence the to release the encapsulated dye. Fig. 7 illustrates that during the increase of liposomes concentration from 1 to 10 g/l, marked changes in the liposome size were observed. Thus by increasing liposome concentrations from 1 to 4 g/l, the  $K/S$  value at different temperatures (60, 80 and 95 °C) generally increased especially at 95 °C. However, when raising the liposome concentrations from 4 to 10 g/l,  $K/S$  value generally decreased.

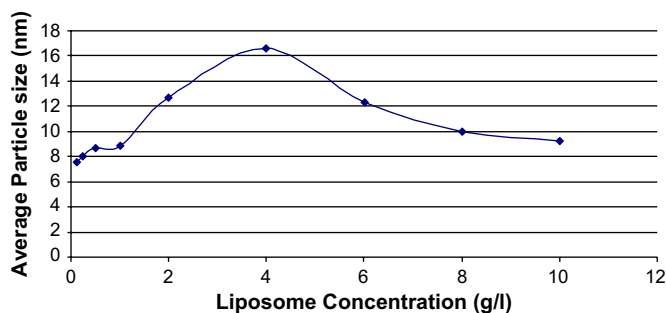


Fig. 4. Effect of various liposome concentrations on the particle size of nano-encapsulated C.I. Reactive Red 194 emulsion.

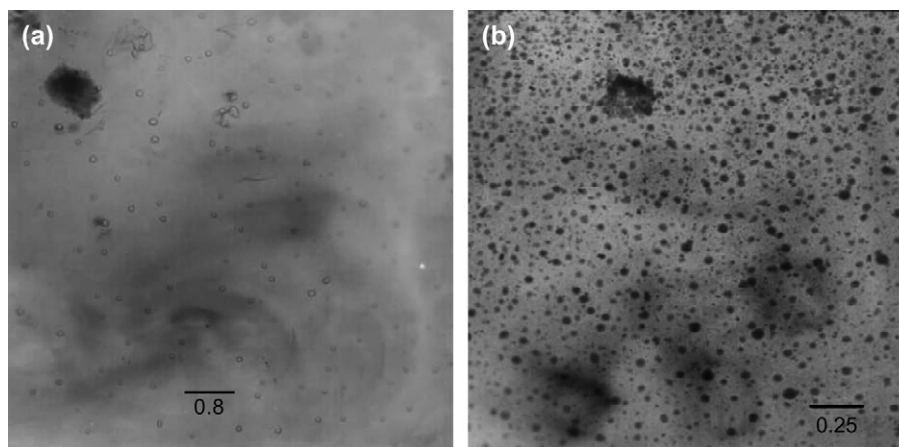


Fig. 6. Transmission electron microscopy images of: (a) 4 g/l acetylated AIF liposome emulsion (with no dye) as nanospherical vesicles, with magnification 12,500 $\times$  (b) 4 g/l acetylated AIF liposome emulsion with encapsulated heterobifunctional reactive dye, with magnification 40,000 $\times$ .

These results indicated clearly that at lower liposome concentrations up to 4 g/l, the encapsulated dye increased, however, concentrations in case of above 4 g/l the liposomes shell or membrane may be thick-walled, or possibly became multilayer that allowed lower amounts of dye to be encapsulated in the liposome vesicle. In other words, the liposomes containing dye are more resistant to release the dye even after adding Triton X-100, i.e. more stable [17–20].

It can be concluded that 4 g/l liposomes was an optimum concentration and therefore it was advisable to study the influence of different time periods (10–180 min.) on this optimal liposome concentration at constant temperature of 95 °C.

It is clear from previous studies [3,17] that if the dyeing time increased, the  $K/S$  values and final dyebath exhaustion also increased, showing that liposome stability decreased with time. Owing to the fact that longer dyeing time and lower exhaustion percent and  $K/S$  values are not feasible from the economic point of view, an alternative method was adopted using Triton X-100 after only 30 min from the beginning of the dyeing process to destroy the liposome and to save time and energy while  $K/S$  value increased as represented in Fig. 8. Consequently, it was found that better results were obtained when using 4 g/l liposome at 95 °C for 60 min dyeing time.

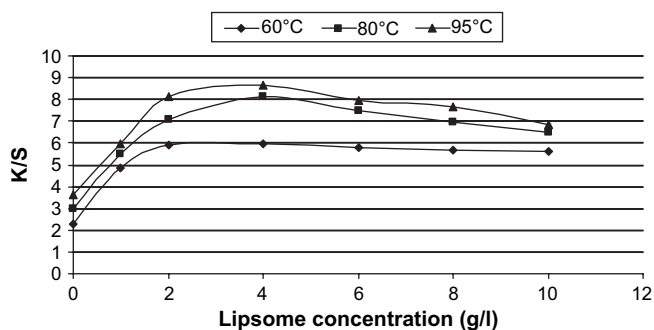


Fig. 7. Effect of different dyeing temperatures and different concentrations of liposome on the colour strength of dyed wool fabrics with C.I. Reactive Red 194.

### 3.7. Effect of salt concentration

As it was reported previously that addition of sodium sulphate salt in wool dyeing is necessary in the case of using high affinity acid or reactive dyes as it retards the dye migration and thus achieving better level dyeing. Fig. 9 shows the influence of salt concentration (0–30% owf) on LERD and the conventional (non-liposome) reactive dye (CRD). However,  $K/S$  values indicated that negligible increase from 8.46 to 8.64 was observed as the salt concentration increased from 0 to 10% owf, however, it decreased by further increase in salt concentration from 15 to 30% owf. Therefore, 10% owf salt concentration was considered as an optimum concentration that gave  $K/S$  value of 8.64 compared to 3.64 in the conventional (non-liposome) wool dyeing. These results emphasized that dyeing wool with LERD reduced the amounts of electrolyte used.

### 3.8. Effect of dyeing temperature

Applying the optimum salt concentration (10% owf) on the dyeability of wool fabrics at different dyeing temperatures (40–95 °C) using LERD and CRD method, the results obtained are recorded in Fig. 10. It was found that the  $K/S$  values increased with increasing the dyeing temperature in both cases

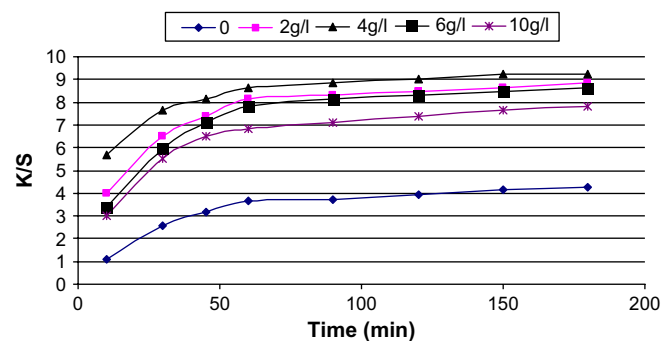


Fig. 8. Effect of different dyeing times and concentrations of liposome on the colour strength of dyed wool fabrics with C.I. Reactive Red 194.

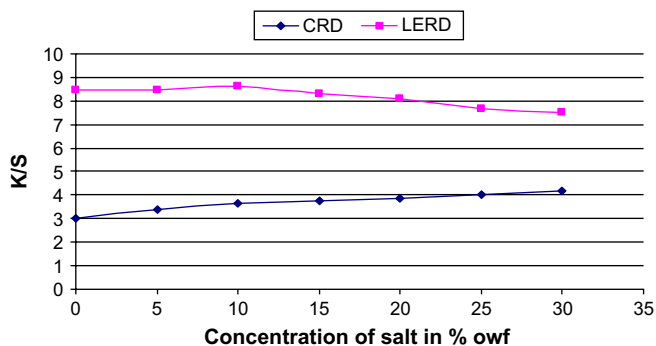


Fig. 9. Effect of salt concentrations on the colour strength of dyed wool fabrics with C.I. Reactive Red 194 with and without liposome nanoencapsulation.

of LERD and the conventional wool dyeing process. However, the colour strength of the dyed wool fabric with LERD showed superior values to the conventional dyeing reactive dye at different temperatures especially at 95 °C. The higher temperature of 95 °C enhanced the distribution of the dye released gradually from the liposome to the wool fibre as indicated from the values of  $K/S$ .

### 3.9. Effect of dyeing time

Wool dyeing process with both CRD and LERD at different time intervals under the optimum conditions (4 g/l liposome and 10% owf salt at 95 °C) showed that exhaustion and total fixation percent in both cases, increased as the dyeing time increased from 10 to 180 min as shown in Fig. 11. In case of LERD wool dyeing process, both the final dyebath exhaustion and total fixation percent were achieved after 60 min of the dyeing time. It seems that equilibrium is reached after 120 min of dyeing process when using LERD dyeing method. In contrast, the equilibrium did not reach even after 180 min in case of the CRD which may be due to the dye aggregation on the surface of the fibre as a result of the long dyeing time. Accordingly, LERD wool dyeing process improved the exhaustion and total fixation efficiency. Thus, reaction parameters that led to the optimum pure wool dyeing conditions can be summarized as follows: 4 g/l acetylated AIF with LERD, 10% owf salt at 95 °C for 60 min and Triton X-100 was added after 30 min from the beginning of the dyeing process.

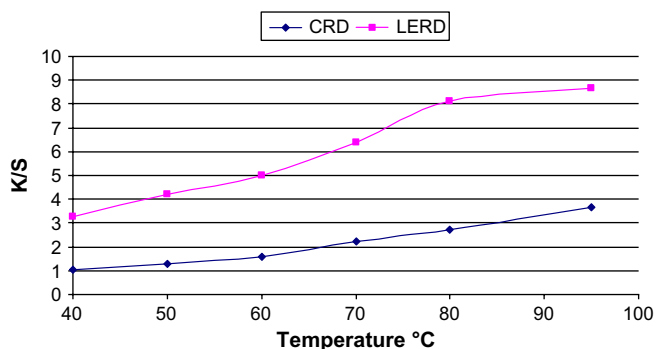


Fig. 10. Effect of dyeing temperature on the colour strength of dyed wool fabrics with C.I. Reactive Red 194 with and without liposome nanoencapsulation.

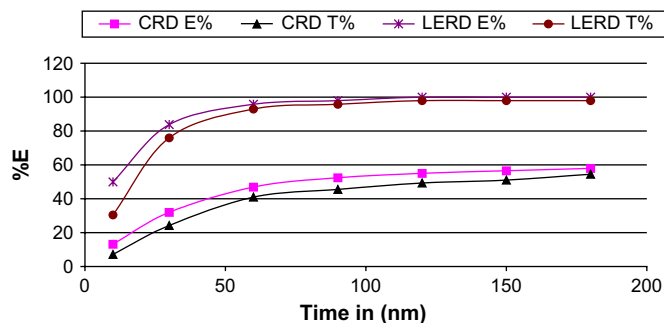


Fig. 11. Effect of dyeing time on the exhaustion (%E) and total fixation (%T) of dyed wool fabrics with C.I. Reactive Red 194 with and without liposome nanoencapsulation.

### 3.10. Application of LERD on dyeing wool blend fabrics

After achieving the optimum conditions of pure wool dyeing with LERD, it was feasible to apply the same optimum dyebath conditions on wool blend fabrics in comparison with pure wool using one-bath technique as shown in Table 2. The selected and available wool/blend fabrics in this work were polyester/wool (55/45%) the most popular blend, cotton/wool (70/30%) and acrylic/wool (67/33%). The results recorded in Table 2 indicated that polyester/wool blend fabric exhibited the maximum values of  $K/S$ , exhaustion percent ( $E\%$ ) and total fixation percent ( $T\%$ ). This can be due to the fact that the liposomes of LERD were bound with the nucleophilic amino and thiol groups present in wool fibre; however, they could not bind with the non-ionic polyester fibre [21].

Although cotton/wool blend fabrics responded to LERD dyeing, yet the liposome dyeing efficiency was higher in pure wool fibres than in cotton fibres. These results can be attributed to the easier bonding of liposomes with the amino and thiol groups in wool fibre than with the free hydroxy groups in cotton fibre. In case of acrylic/wool blend, the results indicated that LERD prefer to form stable bonds with wool fibre than with acrylic fibres. This may be due to the repulsion force between the negative charge of the acrylonitrile groups on the acrylic fibre and the reactive group of the reactive dye and liposome. It is clear from Table 2 that conventional reactive dyeing of wool exhibits no satisfactory results as the LERD method.

Table 2

Effect of dyeing wool blend fabrics with C.I. Reactive Red 194 on the colour strength in the presence and absence of liposome

| Kind of fabrics | Blend ratio in % | CRD         |       |       | LERD        |       |       |
|-----------------|------------------|-------------|-------|-------|-------------|-------|-------|
|                 |                  | $K/S$ value | $E\%$ | $T\%$ | $K/S$ value | $E\%$ | $T\%$ |
| Wool            | 100              | 3.64        | 50.50 | 39.9  | 8.64        | 96.0  | 90.0  |
| Polyester       | 100              | 0           | 0     | 0     | 0           | 0     | 0     |
| Cotton          | 100              | 3.29        | 43.0  | 38.0  | 5.29        | 77.60 | 65.5  |
| Acrylic         | 100              | 0           | 0     | 0     | 0           | 0     | 0     |
| Polyester/wool  | 55/45            | 1.637       | 30    | 23.13 | 3.91        | 80.33 | 64.92 |
| Cotton/wool     | 70/30            | 1.795       | 48.0  | 38.59 | 2.54        | 78.0  | 70.0  |
| Acrylic/wool    | 67/33            | 1.220       | 26.0  | 23.83 | 1.88        | 63.72 | 58.50 |

Meanwhile, the efficiency of LERD on dyeing different wool/blend fabrics can be arranged in the following order: polyester/wool > cotton/wool > acrylic/wool.

#### 4. Conclusion

From the results obtained from the present work the following conclusions can be derived out: chemically modified AIF soybean phospholipids mixture was capable of forming liposomes for encapsulating reactive dyes, which were applied in dyeing of pure wool as well as wool blend fabrics and substantially act as a levelling, exhausting and fixing agent. The liposomes of AIF of soybean phospholipids can form nanoemulsion as indicated from particle size measurement and this emulsion was very effective in encapsulating the dyes physically in the liposome vesicles. Thus, using the nanoencapsulation method with acetylated AIF phospholipids proved to be superior to the conventional wool dyeing method. Meanwhile, the encapsulated dye can be slowly released towards the wool fibres to achieve better dye uptake. Liposomes (as non-contaminant biological materials) are known to be easily biodegradable than those conventionally synthesized auxiliaries and can also be considered as an additional advantage for this novel technology. In addition, the application of this technology resulted in saving energy by reducing time and temperatures more than those needed in the conventional wool dyeing method, avoiding the use of any synthetic auxiliaries. Future work in liposome technology will be extended in the field of textiles using different modified phospholipids mixtures to produce tailor-made emulsifiers with specific properties.

#### Appendix

|  |                  |
|--|------------------|
| 1. Soybean lecithin                          | SBL              |
| 2. Acetone insoluble fraction                | AIF              |
| 3. Microencapsulated dye                     | MS               |
| 4. Phosphatidylcholines                      | PCs              |
| 5. Liposomes encapsulating reactive dye      | LERD             |
| 6. Phosphatidylcholine                       | PC               |
| 7. Phosphatidylethanolamine                  | PE               |
| 8. Colour strength                           | <i>K/S</i> value |
| 9. Exhaustion %                              | <i>E</i> %       |
| 10. Fixation %                               | <i>F</i> %       |
| 11. Total fixation %                         | <i>T</i> %       |
| 12. Lysophosphatidylcholine                  | Lyso-PC          |
| 13. Phosphatidic acid                        | PA               |
| 14. Phosphatidylinositol                     | PI               |
| 15. Lysophosphatidylethanolamine             | Lyso-PE          |
| 16. Conventional (non-liposome) reactive dye | CRD              |

#### References

- [1] Nelson G. Application of microencapsulation in textiles. *International Journal of pharmaceutics* 2002;242:55–62.
- [2] Baptista ALF, Goutinho PJG, Real Oliveira MECD, Rocha Gomes JIN. Lipid interaction with textile fibres in dyeing conditions. *Progress in Colloid and Polymer Science* 2004;123:88–93.
- [3] Rocha Gomes JIN, Genovez MC, Hrdina R. Controlling exhaustion of reactive dyes on wool by microencapsulation with liposomes. *Textile Research Journal* 1997;67(7):537–41.
- [4] De La Maza A, Goderch L, Manich AM, Marti M, Parra JL. Optimizing a wool dyeing process with an azoic 1:2 metal complex dye using commercially available liposomes. *Textile Research Journal* 1998;68(9):635–42.
- [5] Goderch L, Manich AM, Marti M, De La Maza A, Parra JL, Serra S. Complementary study of optimizing a wool dyeing process with commercially available liposomes. *Textile Research Journal* 1999a;69(10):789–90.
- [6] Goderch L, Marti M, De La Maza A, Manich AM, Parra JL, Serra S. Potential of liposomes to aid the quest for cleaner dyeing. *Wool Record*; 1999b. Dec. 26.
- [7] De La Maza A, Manich AM, Goderch L, Parra JL. Multilamellar liposomes including cholesterol as carriers of azobenzene disperse dyes in wool dyeing. *Textile Research Journal* 1995;65(3):163–70.
- [8] De La Maza A, Parra JL. Unilamellar lipid bilayers as vehicles of azo disperse dyes on wool. *Textile Research Journal* 1994;64(5):255–61.
- [9] Carrion Fité FJ. Dyeing polyester at low temperatures: kinetics of dyeing with disperse dyes. *Textile Research Journal* 1995;65(6):362–8.
- [10] New RRC. In: New RRC, editor. *Liposomes – a practical approach*. NY: Oxford University Press; 1990. p. 221–52.
- [11] Ziegelitz Rudiger. Lecithin processing possibilities. *Inform* 1995;6(11): 1224–30.
- [12] Nzai JM, Proctor A. Phospholipids determination in vegetable oil by thin-layer chromatography and imaging densitometry. *Food Chemistry* 1998;63(4):571–6.
- [13] Hollo J, Peredi J, Ruzics A, Jeranek M, Erdelyi A. Sunflower lecithin and possibilities for utilization. *Journal of the American Oil Chemists' Society* 1993;70(10):997–1001.
- [14] Judd B, Wysecki G. *Colour in business science and industry*. 3rd ed. New York: John Wiley and Sons, Inc.; 1975.
- [15] Lewis DM, Renfrew AH, Siddique AA. The synthesis and application of new reactive dye based on disulfide-bis-ethylsulfone. *Dyes and Pigments* 2000;47:151–67.
- [16] Lewis DM, Shao JZ. A new approach to the dyeing of silk with sulpha-toethylsulphone dyes. *Journal of the Society of Dyers and Colourists* 1995;111:146.
- [17] De La Maza A, Parra JL, Bosch P, Coderch L. Large unilamellar vesicle liposomes for wool dyeing: stability of dye–liposome systems and their application on untreated wool. *Textile Research Journal* 1992;62:406–13.
- [18] Lichtenberg D, Robson J, Dennis EA. Solubilization of phospholipids by detergents. *Biochimica et Biophysica Acta* 1983;737:285–304.
- [19] De La Maza A, Parra JL. Vesicle–micelle structural transition of phosphatidylcholine liposomes and Triton X-100. *Biochemical Journal* 1994;303:907–14.
- [20] De La Maza A, Parra JL, Goderch L, Bosch P. Phosphatidylcholine/cholesterol liposomes as vehicles for anthraquinone disperse dyes in wool dyeing. *Journal of the Society of Dyers and Colourists* 1995;111:30–5.
- [21] Marti M, Goderch L, De La Maza A, Manich A, Parra JL. Phosphatidylcholine liposomes as vehicles for disperse dyes for dyeing polyester/wool blends. *Textile Research Journal* 1998;68(3):209–18.