Microencapsulation of Disperse Dye Particles with Nano Film Coating Through Layer by Layer Technique

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ABSTRACT: Throughout the past ten years, comprehensive understanding of fundamental and applied research has focused on functional coating and specifically on microencapsulaion. In this study, weak polycation poly(allylamine hydrochloride) and strong polyanion poly(sodium styrene sulfonate) were used for fabrication of nano film through layer by layer technique on the surface of disperse dye particles. Then micron-sized particles were surrounded by poly(urea formaldehyde) using *in-situ* polymerization. Chemical structure, surface morphology, and size distribution of these novel microcapsules were characterized by Fourier transform infrared spectrometry, differential scanning calorimetry, optical microscopy, and scanning electronic microscopy. Size and surface morphology of the microcapsules can be optimized by selecting proper weight ratio of urea to formaldehyde and core to shell material type, and amount of surfactant and agitation rate. This technology demonstrated good capability in several applications in textile industry, such as dying fabrics because of saving huge amount of water and showing slow-release property of dye without using dye assistant agents. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 119: 586–594, 2011

Key words: microencapsulation; disperse dye; layer by layer self assembly; polyelectrolyte; poly(urea formaldehyde); *in-situ* polymerization

INTRODUCTION

Nowadays, in the modern era of technology the value of coating has been extensively enlarged. The process or the resultant dried continuous or discontinues film which is applied on to the surface is called coating. For special performance, coating which is organic or inorganic could be applied as multilayer system. In this kind of coating, each layer is influenced by other layers, while it keeps its specific functionality.[1] Additional functionalities, beside decorative and protective is defined as the term of "functional coatings".[2]

Throughout the past 10 years, one of the applications of the functional coatings which has been explored widely by the agricultural,[3,4] pharmaceutical,[5–7] food,[8,9] and textile science and industry[10–31] is microencapsulation technology. The process of surrounding micron-sized particles of solids, droplets of liquids and gasses in an inert shell is defined as the microencapsulation process. This technology is used for protection, controlled release, and compatibility of the core materials.[2,32]

The developing application of microencapsulation in textiles is restricted to pigments,[12,13] dyes,[14–29] fragrances,[30] and phase-change materials.[31] Using of microencapsulation technology in textile has been limited because of two reasons: lack of awareness about this technology and being relatively expensive process. Microencapsulation of the pigments, such as microencapsulation of ultramarine[12] and copper phthalocyanine[13] has been previously reported. Microencapsulation of dyes, such as carbonless copy paper,[14,15] metal complex dyes in dyeing wool and wool blends[16–21] and different class of dyes like disperse dyes,[22–24] acidic dyes,[25] and azo dyes[26–28] are available. A review article relating the photosensitive color producing microcapsules has been also published.[29]

In this study, we used disperse dye particles as a core material. Disperse dyes are low molecular weight and nonionic molecules. Their chemical constitutions are monoazo or anthraquinone derivatives. They are slightly soluble in water as a result of both presence of polar substituent in dye molecule, such as hydroxyethylamino groups ($NHCH_2CH_2OH$), and their relatively low molecular weight.[33]

One of the most powerful tools for the fabrication of multilayer flat thin film through layer by layer technique is self assembly of oppositely charged polyelectrolyte. During the past few years scientists

produced stimuli responsive microcapsules by applying this technique in microencapsulation technology.[34] Environmental changes can affect these kinds of microcapsules, hence physical or chemical modification will occur on the surface of microcapsules. In this study, we used weak polycation poly(allylamine hydrochloride) and strong polyanion poly(sodium styrene sulfonate) for fabrication of thin film through layer by layer technique on the surface of disperse dye particles. Then the resultant micron-sized particles were surrounded by poly(urea formaldehyde) using *in-situ* polymerization.

In this work, polymerization of urea formaldehyde occurs separately in the interface of the core material which is polyelectrolyte coated disperse dye. No active agent for the shell polymerization exists in core material.[2,32] The initial shell materials form a liquid like solution called prepolymer. As molecular weight increases, the hard, non-sticky, and stable protective shell is formed on the surface of the core as result of deposition and crosslinking of urea formaldehyde. Anionic and cationic surfactants were used to facilitate deposition of poly(urea formaldehyde) on surface of the core.

Therefore, for improving the property and quality of dyes, instead of altering the chemical structure of dyes, coating the dye particles via microencapsulation techniques by means of layer by layer self assembly technique have been used. This technique is easy to assemble rather than using different active components and creating significant changes on dye properties. Since, disperse dye is insoluble in aqueous solutions, introduction of hydrophilic shell could be valuable in some applications.

Cellulose acetate and all synthetic fibers can be colored using disperse dyes. The dyeing adsorption isotherm is Nernst isotherm, in which the variation of absorbed dye concentration against its solution concentration is linear to the saturation point of dye in water. Further than this point; the dye concentration in fiber will be in equilibrium with the saturated aqueous solution, while the saturated aqueous solution is in equilibrium with solid dye particles that are disperse in solution using surfactants. Microencapsulation technology provides a proper release characteristic for monomolecular dyes. Consequently, the slop of the Nernst isotherm graph will be optimized in the required temperature. On the other side, surfactants decrease the dying exhaustion because of the apparent water solubility that they provide and they cause pollution in waste waters. Therefore their substitution with microcapsules containing disperse dye could be very advantageous to have environmental friendly textile dyeing process. In the textile industry, most concerning issues focus on waste water, especially when auxiliary agents, such as penetrating agents, dispersing agents, etc., should be added to the dyestuffs and also the dye bath recovery and reusing after dyeing procedure. So that, huge amount of water can be saved. Preventing from both dust formation (while powder form is used) and aggregation and bulk formation of dyes in dye bath are their lateral advantages. In some cases where the mixture of the dyes is desirable, dyes particles physically repel each other so microencapsulation technology could be used to overcome this problem.

In another point of view, microcapsules containing disperse dyes have potential to be used in textile printing where novel multicolor designs, such as duplex multilayer fabrics are required. In these situations, transfer printing papers can be reusable as result of the control release properties of semipermeable microencapsulated disperse dye. Finally shell materials, like poly(urea formaldehyde), are able to keep moisture on the fabric surface during printing process, as is used in conventional textile printing formulation.

For investigation of chemical structure, surface morphology and size distribution microcapsules were characterized by Fourier transform infrared spectrometry (FTIR), optical microscopy (OM), differential scanning calorimetry (DSC) and scanning electronic microscopy (SEM).

EXPERIMENTAL

Methods

C.I. Disperse Blue 1 which purchased from Sunny Chemical (China) was used as core material. Weak polycation poly(allylamine hydrochloride) and strong polyanion poly(sodium styrene sulfonate), which were used as first shell layer were purchased from Fluka Chemie GmbH (Germany). Urea and formaldehyde (37 wt %) used as second shell layer, were obtained from Fluka. Sodium lauryl sulfate and cetrimonium bromide are used as cationic surfactant, respectively. Sulfuric acid (5 wt %) solution was prepared to reduce the pH of emulsion. Scheme 1 shows the chemical structures of C.I. Disperse Blue 1, poly(allylamine hydrochloride), poly(sodium styrene sulfonate), urea, and formaldehyde.

Preparation of microcapsules

The first layer on the dye particles was formed by poly(sodium styrene sulfonate) as follow: 2 mL of 2 mg mL^{-1} aqueous solution of poly(sodium styrene sulfonate) in 0.5 M NaCl was sloped to a 100 mL beaker. Then disperse dye particles were added to the solution [Fig. 1(a)]. The suspension was left for 20 min with infrequent shaking. After three times centrifuging process for removing the excess amount
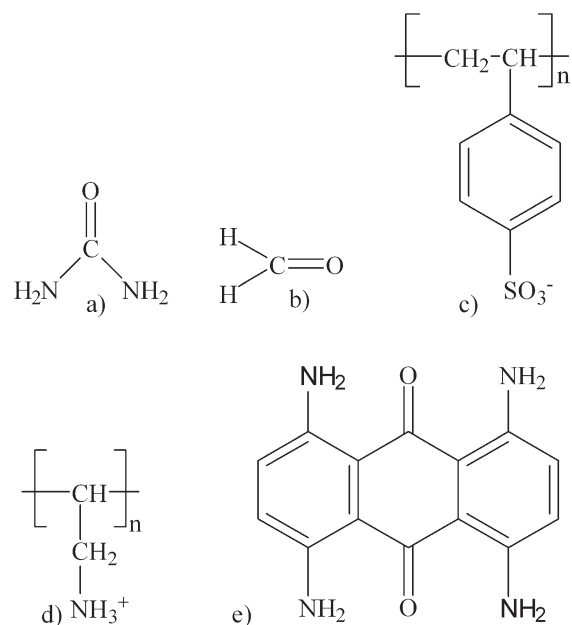
Scheme 1 Chemical structures of materials: (a) urea, (b) formaldehyde, (c) poly(sodium styrene sulfonate), (d) poly(allylamine hydrochloride) and (e) C.I. Disperse Blue 1.

Figure 1 Schematic of the microencapsulation process.

of poly(sodium styrene sulfonate), the suspension was washed with 0.5 M NaCl [Fig. 1(b)]. The procedure of deposition of second layer, using 2 mg mL^{-1} solution poly(allylamine hydrochloride) containing 0.5 M NaCl, was the same as the first layer [Fig. 1(c,d)].

In the next step, urea, formaldehyde, and deionized water were mixed at room temperature with stirrer at rate of 200 rpm. After 30 min while the temperature was increased to 65–70°C the transparent urea formaldehyde prepolymer solution was obtained. Aqueous solution of surfactant (100 mL of 2 wt %) was added to prepolymer solution [Fig. 1(e)]. The slow stream of disperse polyelectrolyte coated dye particles was added to form emulsion and the rate of stirrer was increased. After 30 min the pH of emulsion was decreased slowly to 3 using sulfuric acid solution (5 wt %) and the agitation rate was decreased. This step of preparation has taken
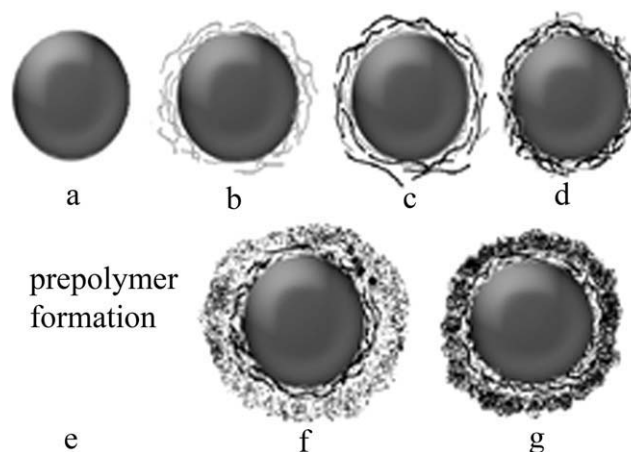
5 h to complete. In this step the pH of solution must be continually controlled. At the end of the process a slurry suspension was obtained [Fig. 1(f,g)]. Table I shows the processing conditions of microcapsules containing disperse dye. The suspension was rinsed with deionized water and methanol and filtered. At the end, to inhibit from the formation of cohesion between microcapsules, we equilibrated them for 1 min under ultrasonic waves. Figure 2 shows the schematic of the reactor which was used for coating poly(urea formaldehyde) on the surface of polyelectrolyte coated disperse dyes.

Characterization method

Chemical structure of samples was identified using FTIR (BRUKER EQUINOX 55) spectrometer. For deploying IR spectroscopy of crushed solid samples, e.g., in this study, sample is placed onto the high purity salt plates. For preparing the dye and microencapsulated dye sample, we mixed crushed high purity KBr with our solid sample and formed a translucent pellet to investigate directly. We used KBr to decrease the crystal reflection of our samples.

TABLE I
Microcapsules Prepared at Different Processing Conditions

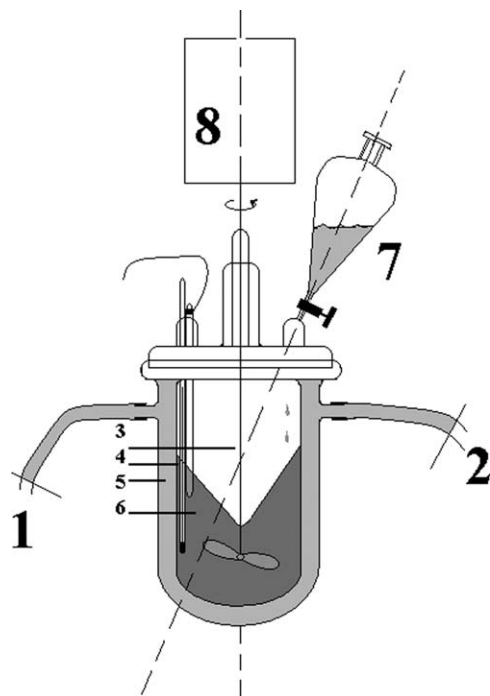| Sample	Surfactant type	Agitation rate (rpm)	Weight ratio of urea toward 37 wt % formaldehyde	Weight ratio of core material toward shell material
1	Anionic	500	1–2	1–100
2	Anionic	500	1–4	1–100
3	Anionic	500	1–2	1–20
4	Anionic	500	1–2	3–4
5	Anionic	100	1–2	1–100
6	Anionic	800	1–2	1–100
7	Cationic	500	1–2	1–100

Figure 2 Schematic of reactor: (1) and (2) tubes for insertion and exertion of hot or cold liquid (in this work water) to control temperature of system (3) stirrer (4) thermometer and pH meter (5) three-necked flask (6) continuous phase (7) decanter for controlling drop wise rate (8) stirrer electric motor.

The thermal properties were analyzed by using differential scanning calorimetry (NETZSCH DSC, 200 F3) at a heating rate of 10°C/min from 25 to 400°C in a nitrogen atmosphere.

Optical microscopy (ZEISS-JENAPOL) was used to investigate the size distribution of prepared microcapsules. Scanning electronic microscope (SEM -Cambridge-S360) with Image analyzing system

(SIS) used to observe surface morphology of the microcapsules. Each sample was fixed on standard sample holder and sputter coated with gold using coater.

RESULTS AND DISCUSSION

Chemical structure of microcapsules containing disperse dye

The infrared spectroscopy was used to investigate chemical structure of the prepared polymers. Figure 3 indicates FTIR spectra of prepolymer, dye, urea, and microcapsules of sample number 1 containing disperse dye. Doublet bands at 3445 and 3355 cm^{-1} are presented by the FTIR spectrum of urea. These absorption bands can be associated with the NH groups of urea. As it can be seen, polycondensation reaction between urea and formaldehyde were proved by the absence of absorption band owing to urea at 2806 and 2640 cm^{-1} and manifestation of absorption peak of poly(urea formaldehyde), which is assigned at 3707–3050 (NH and OH), 1649 ($\overset{\text{O}}{\overset{\|}{-\text{NH}-\text{C}-\text{NH}-}}$), 1544 ($\overset{\text{O}}{\overset{\|}{-\text{C}-\text{NH}-}}$) and 1027 ($-\overset{|}{\text{CH}}-\text{O}-\overset{|}{\text{CH}}-$) cm^{-1}. On the other hand, the absorption peaks of 1556, 1035, and 630 cm^{-1} are appeared in both microcapsules and dyes spectra. This indicates that the core content has been embraced with poly(urea formaldehyde). This embracing phenomenon will not be achieved in all encapsulation conditions. Basically, interfacial tensions between core material and medium phase, core material and shell material, and also medium phase and shell material, are the basic criteria that undertake the final equilibrium state.
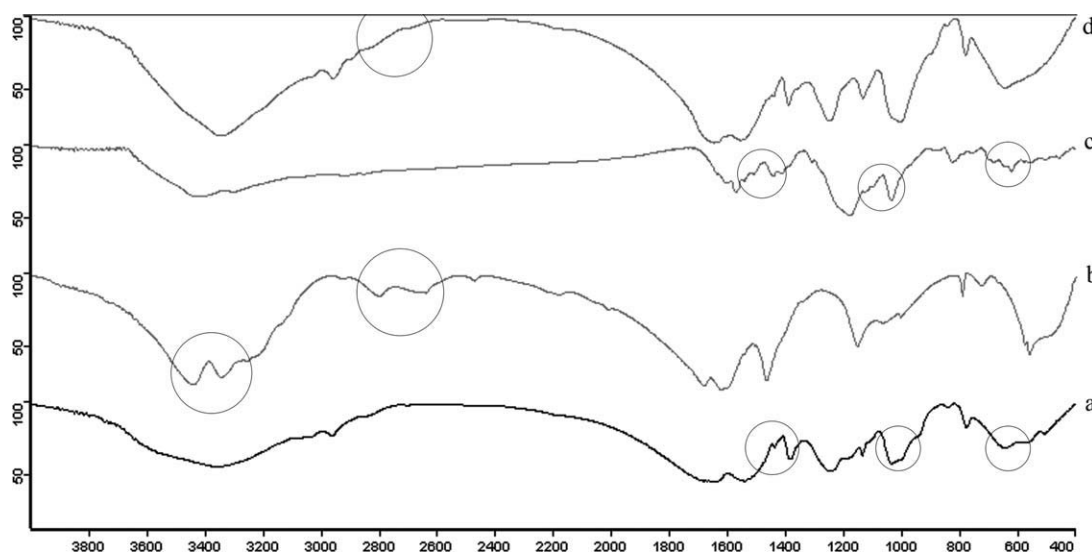

Figure 3 FTIR spectra of (a) microcapsule containing disperse dye, (b) urea, (c) disperse dye, and (d) prepolymer.
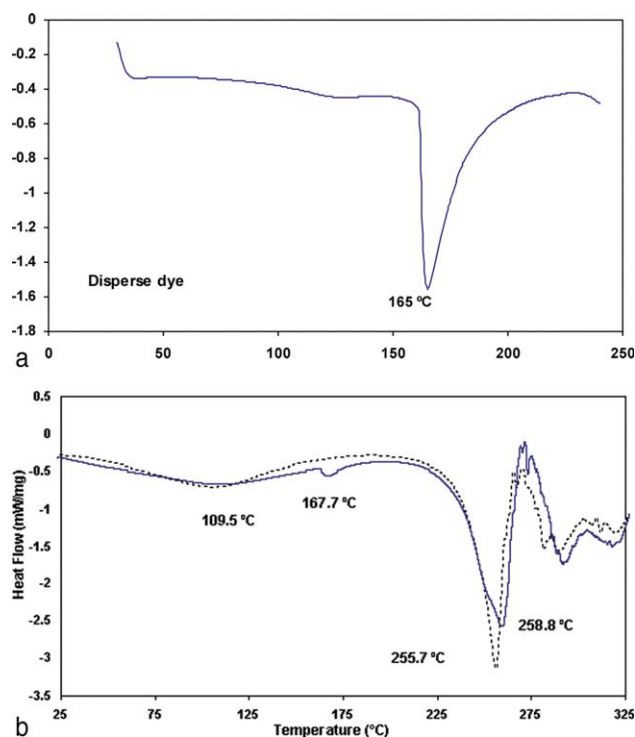
Figure 4 (a) Dispers dye, (b) The DSC thermograms of hollow microcapsules (Dashed line) and microcapsules containing dye (Solid line). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Thermal property of microcapsules containing dye

The DSC thermogram of disperse dye hollow microcapsules (dash line) and microcapsules containing dye (solid line) are shown in Figure 4(a,b), respectively. Two endothermic peaks at 105.5 and 255°C appear in the DSC thermogram of microcapsules. The first endothermic peak below 109°C is related to the evaporation of water and free formaldehyde.[35,36] The second one at temperatures about 250°C is due to the decomposition of polyureaformaldehyde. Dye exhibits a sharp endothermic peak at 167°C. SEM micrographs and the dye endothermic peak indicate that most of the dye particles were loaded in UF microcapsules.

Surface analyses of microcapsule containing disperse dye

To achieve the desired properties of microcapsules, their surface morphology would be evaluated. First layer of polyelectrolyte applied on dye particle through combination of electrostatic, hydrophobic, and van der Waals interactions. The dye which we used was uncharged and poly(sodium styrene sulfonate) was coated on the particles at the first. With the adsorption of each layer, the overall charge of the surface will be reversed which expedites the

deposition of next layer. Since NaCl decrease the binding site for each chain, thus the deposition of polyelectrolyte on the surface will be increased. Therefore NaCl is added to polyelectrolyte dipping solution.[2] Polyelectrolytes are soluble in polar solvents like water and they possess several oppositely charged counter ions. Segmental hydration and free energy of mixing are the main reasons for their solubility and phase behavior. The higher charged density and more flexibility will provide more expansion in water than nonionic polymers, at low ionic strength. Temperature, PH, or electrolytes will provide a substrate for modifying molecular structure which allows large conformation charge. Therefore this layer undergoes reversible swelling by changing the pH. The main concern in layer by layer technique is removing the excess amount of polyelectrolyte from the polyelectrolyte dipping solution before the next deposition to prevent the development of polyelectrolyte complexes in bulk solution. There are several ways to remove this excess amount such as centrifugation, washing, and resuspension. In this article, we used this centrifugation method which is widely used for layer by layer assembly coating.

In-situ polymerization of urea formaldehyde prepolymer formed three dimensional crosslinked polymers on the outer surface of the particles. The formation of second microcapsule wall is as result of prepolymer surface activity. The prepolymer become augment within the interfaces. In this step, while the prepolymer is still soluble, low molecular weight prepolymer started to deposit on the surface of polyelectrolyte coated dye particle. With adding large amount of acid (pH 2), crosslinking reaction of hydroxymethyl compounds occurred. High molecular weight poly(urea formaldehyde) substance is the result of condensation of urea formaldehyde prepolymer with elimination of water. Since, there is no poly(urea formaldehyde) nanoparticles in the system, smooth and nonporous microcapsule wall is formed. Higher molecular weight prepolymer will precipitate to form poly(urea formaldehyde) nanoparticles and then aggregate or deposit on the surface of the particles. Partial methylolated urea even increases the concentration of reactive prepolymer molecules on periphery regions in consequence of their more surface activity properties. Therefore the polymer condensation proceeds more rapidly in such regions. Obviously, porous and agglomerated microcapsule will be observed in this step.

Figures 5 and 6 show the SEM micrographs of the surface of microcapsules. The roughness of the microcapsules depends on the weight ratio of the urea to formaldehyde which indicates the content of free formaldehyde (sample number 1 and 2). The less the weight ratio causes the surface of microcapsule gets rougher. In common, high concentration of
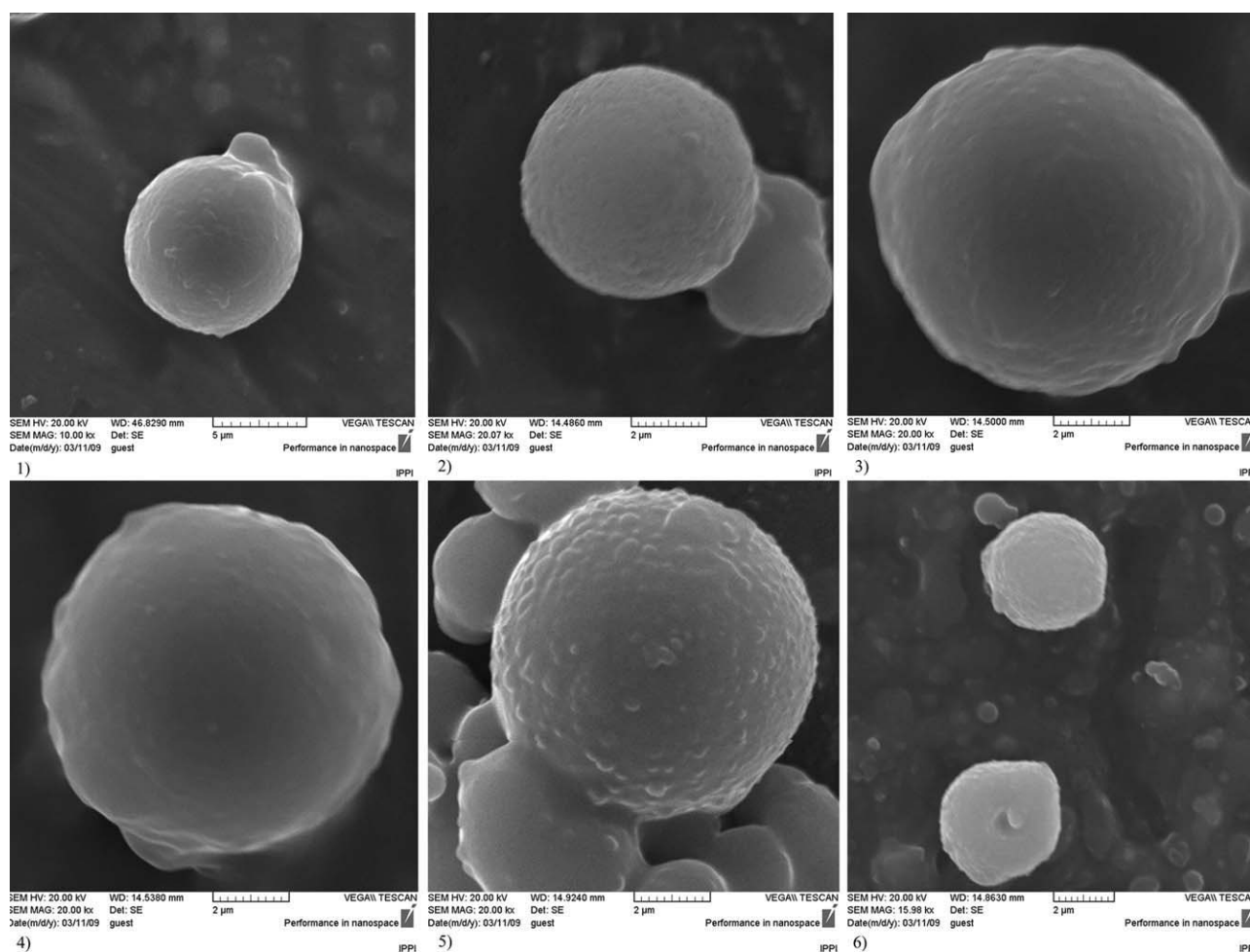
Figure 5 The SEM micrograph of samples number 1–6. Compare to sample number 1 (weight ratio of urea to formaldehyde: 1–2, weight ratio of core material to shell materials: 1–100 and agitation rate: 500), sample number 2 (1–4 weight ratio of urea to formaldehyde), samples number 3 and 4 (1–20 and 3–4 and weight ratio of core material to shell materials, respectively), sample number 5 and 6 (100 rpm and 800 rpm agitation rate, respectively).

free formaldehyde is the result of the low weight ratio of urea to formaldehyde. The agglomerated, rougher and more porous outer layer of the poly-(urea formaldehyde) shell is because of the deposition of the poly(urea formaldehyde) nanoparticles on the surface as a consequence of increasing rate of condensation of urea and formaldehyde in spite of the decrease in the network density of poly(urea formaldehyde). Agglomerated microcapsules as result of poly(urea formaldehyde) nanoparticles deposition are shown in Figure 6.

With enlarging the amount of core materials compare to sample number 1, the smoother microcapsule surface can be achieved (samples number 3 and 4). It could be concluded that both concentration of poly(urea formaldehyde) on each microcapsule and the aggregated splinter of poly(urea formaldehyde) nanoparticles will be lower with increasing the core material of the microcapsules. This reducing in concentration and aggregation of poly(urea formaldehyde) could be as a result of higher surface area of

agglomerated disperse dyes coated with polyelectrolyte in which amount of core materials is high and surfactant concentration is constant.

The agitation rate has a direct effect on the smoothness of the microcapsules surfaces (samples number 1, 5, and 6). Because the deposition rate and compacting the deposits of poly(urea formaldehyde) nanoparticles on the surface of microcapsules decreases with increasing the agitation rate. On the other hand, the poly(urea formaldehyde) nanoparticles conglomeration will be reduced with higher agitation rate (sample number 6).

Investigation on size distribution of microcapsules

The size distribution of core materials plays an important role in enhancing diffusion, permeability or controlled release of microcapsules active agent. Figure 7 shows the size distributions of microcapsules prepared by selecting different agitation rate from 100 to 800 rpm (samples number 5, 1 and 6). With
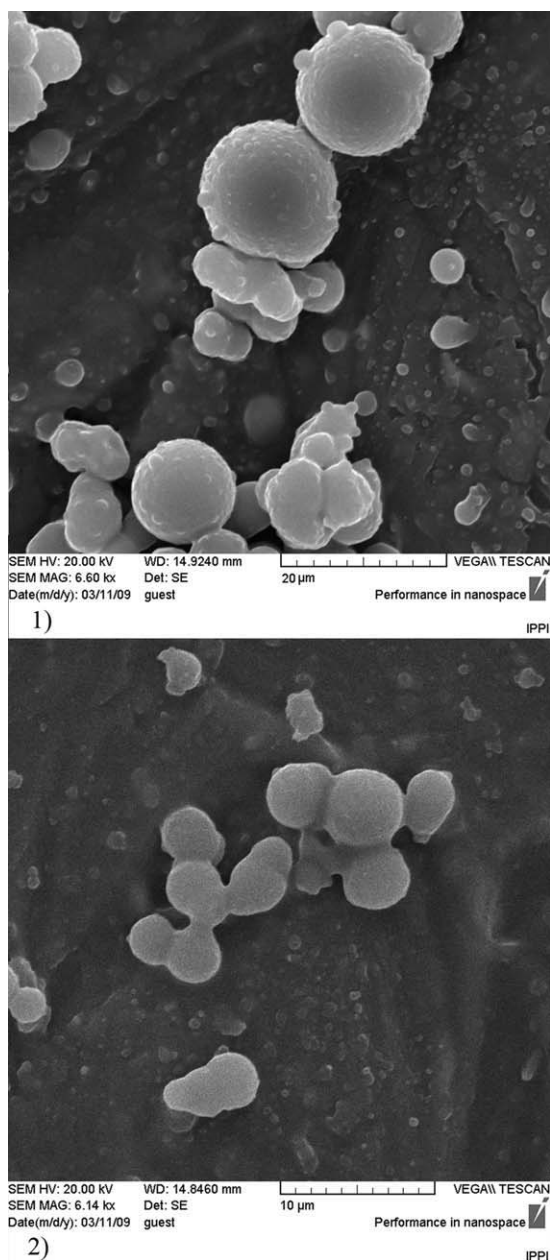
Figure 6 Effect of weight ratio of urea to formaldehyde on microcapsules agglomeration. Sample number (1) and (2) 1–2 and 1–4 weight ratio of urea to formaldehyde, respectively.

increasing the agitation rate and subsequent decrease in the size of the microcapsules, the microcapsule size distribution gets restricted; therefore the smaller microcapsules prevail significantly. The reason is that the high agitation rate causes larger shear stress for the dye particles coated with polyelectrolyte and prevents from dye agglomeration in aqueous solution. Hence, the size of the microcapsules become smaller when the agitation rate is higher, and clearly, the smaller microcapsule diameters rather than lower agitation rate could be observed.

In this work microcapsules with diameters of $\sim$ 3–20 μm were fabricated. Wide range of diameter is for the reason that, near the stirrer blades the turmoil is dominant rather than far from the blades. In this study the average size of diameter is, $\sim$ 10 μm for the sample number 1.

Figure 8 shows the relationship of weight ratio of core to shell materials and microcapsule diameter (sample number 4, 3, and 1). As the weight ratio of core to shell material enlarges, the size of the
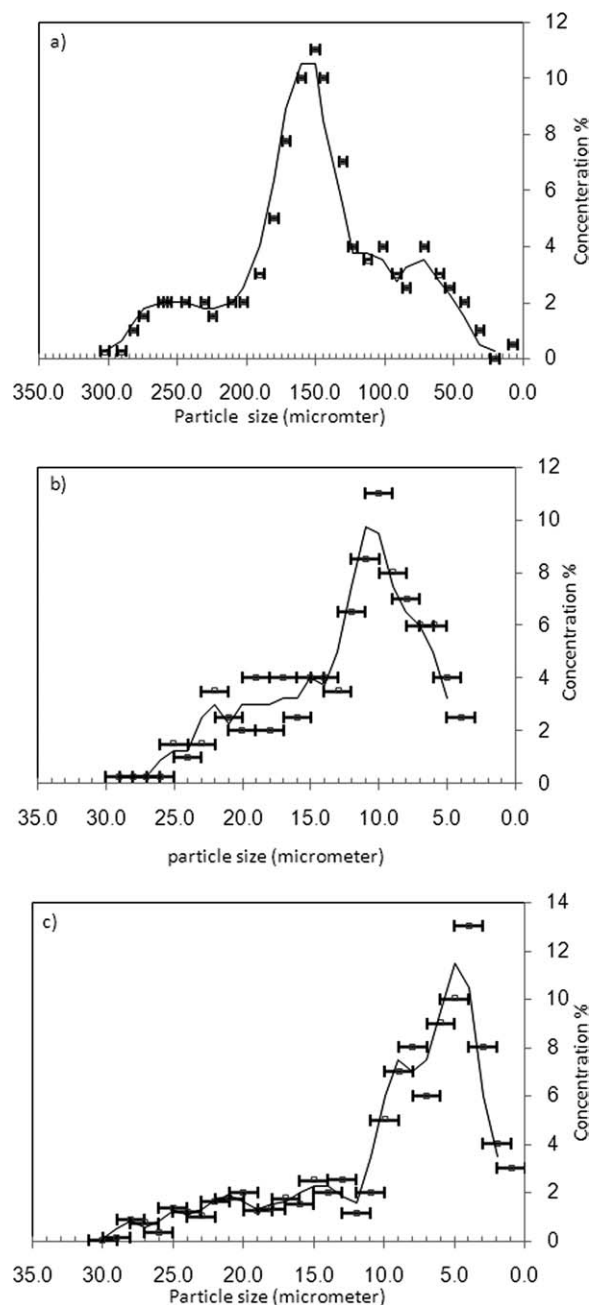

Figure 7 Size distributions of microcapsules (a) sample number 5 (agitations rate: 100 rpm), (b) sample number 1 (agitations rate: 500 rpm), and (c) sample number 6 (agitations rate: 800 rpm).

microcapsules increases. It could be concluded that all the process parameters are constant; the size of disperse dye particles are larger (as a result of dye agglomeration in aqueous solution) when the weight ratio of core to shell material is higher. The decreasing in wall thickness, which is the consequence of the increasing the core materials, can be dominant because it slightly affects the diameter of microcapsule. When the core material diameter significantly increases the larger size of core can be formed, and accordingly, the microcapsule size becomes larger. Moreover, the microcapsules which are prepared by higher weight ratio of core to shell material can be easily fractured due to the thinner wall shell.

The effect of surfactant type on the size distribution of microencapsuls is shown in Figure 9. As it can be seen in the Figures 9 and 7(b) (sample number 1 and 7), the microcapsules containing disperse dye which were prepared with the anionic surfactant possessed smaller average particle size and sharper distribution. Therefore for achieving smaller particle size, using surfactant with negative charge
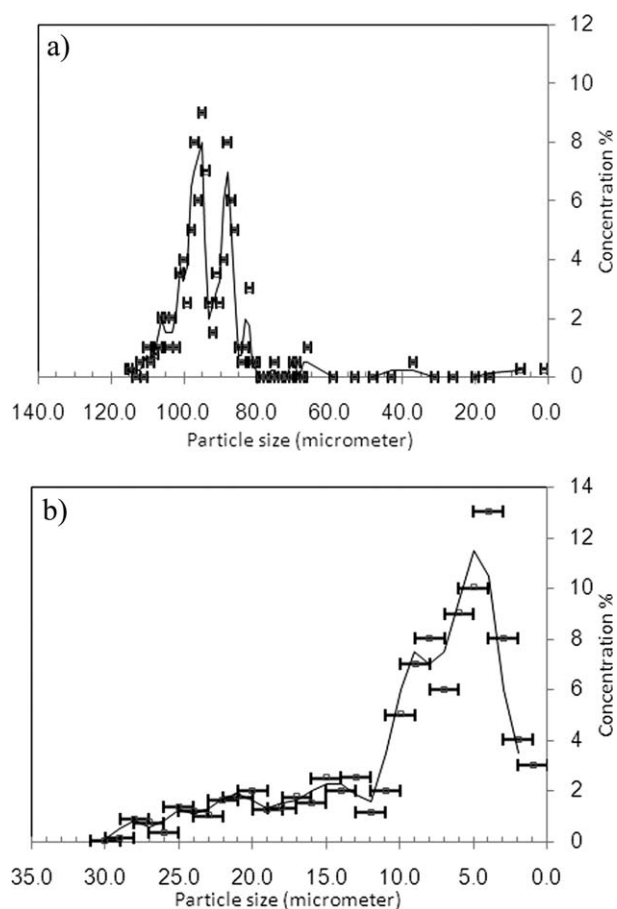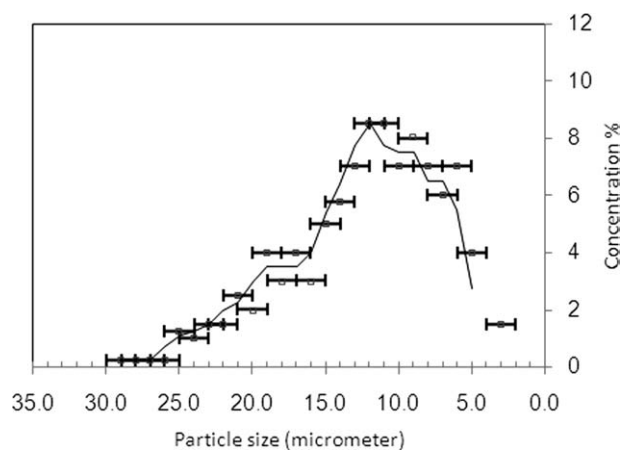

Figure 9 Size distributions of microcapsules prepared by cationic surfactant (sample number 7).

can be more effective because of the electrostatic impediment.

CONCLUSION

In this study microcapsules containing C.I. Disperse Blue 1 which is reasonably anticipated to be a human carcinogen, has been successfully fabricated and different processing parameters have been evaluated. The microcapsules were manufactured in two steps. In the first step disperse dye particles were coated with polyelectrolyte through layer by layer assembly and in the second step poly(urea formaldehyde) nanoparticles were deposited on the surface of microcapsules by reducing the pH to acidic of below 3 at 60–70°C. FTIR spectra indicates that the core content has been embraced with poly(urea formaldehyde). DSC results in addition to SEM pictures showed that the disperse dye was fully engulfed within the microcapsules. The size and the surface morphology of microcapsule could be adjusted by selecting proper weight ratio of urea to formaldehyde and core to shell type of surfactant and agitation rate. During the microencapsulation process, the rough outer surface of microcapsules would be formed in result of rapid deposition of ploy(urea formaldehyde) nanoparticles. Thus, lesser weight ratio of urea to formaldehyde causes the surface of microcapsule gets rougher. SEM micrographs of prepared microcapsules showed that with increasing the agitation rate and core to shell weight ratio separately, the smoother microcapsules surface could be achieved. The relationship between size distribution and agitation rate showed that the size of the microcapsules becomes smaller when the agitation rate is higher. And also the size of the microcapsules increases when the weight ratio of core shell material enlarges. Microcapsules which were prepared

Figure 8 Size distributions of microcapsules (a) Sample number 3 (weight ratio of core material to shell materials: 1–20) and (b) Sample number 4 (weight ratio of core material to shell materials: 3–4).

with the anionic surfactant show smaller average particle size and sharper distribution.

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