INFLUENCE OF MICROENCAPSULATION PARAMETERS ON THE SIZE AND MORPHOLOGY OF MICROCAPSULES BY ECO-FRIENDLY SOLVENT EVAPORATION METHOD ORIENTED TO MEDICAL TEXTILES

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ABSTRACT

Application of microcapsules in medical textile has been studied and commercially developed widely in recent years. The aim of this work was to propose an eco-friendly microencapsulation by solvent evaporation method that can contribute to the manufacture of medical textile products using microcapsules. Quillaja saponin was used as bio-sourced surfactant and ethyl acetate was used as the less toxic, non-halogenated organic solvent. The influences of saponin concentration, stirring speed during emulsification step and the volume of ethyl acetate used to saturate the aqueous phase before emulsification on the microcapsule size and morphology were investigated. The results showed that eudragit microcapsules of ibuprofen with diameter in range from 17 to 34 μm, which were suitable for medical textile applications, were successfully elaborated. The saponin concentration varied from 0.025 to 0.1 wt%. The stirring speed was changed from 700 to 600 rpm. The volume of ethyl acetate used in saturation step was 0, 8 and 12 ml. In the scope of investigation, the saponin concentration and the volume of ethyl acetate used in saturation step did affect the microcapsule size and morphology while the stirring speed did not. The saturation step really helped to reduce the formation of irregular microparticles and to narrow the size distribution, but the microcapsules became more porous, weaker and were deformed significantly by drying in the fabric treatment process.

KEYWORDS

Medical textile; Microcapsule; Solvent evaporation method; Quillaja saponin; Ethyl acetate.

INTRODUCTION

Microcapsules are microparticles in which solid, liquid or gaseous active ingredients (the core) are packaged within the second materials (the shells or the membranes) [1], [2]. With advantages such as the ability of protecting the active ingredients from surrounding environment and controlling the release of active ingredients, microcapsules have been applied in many fields of textile industry including the medical textile [3]–[7].

The size is a very important characteristic of microcapsules because for normal spherical microcapsules, the diameter is in inverse proportion to surface area and therefore, strongly affects the active release rate of active ingredients from microcapsules [1], [5], [8]. The relation between the particle size and release characteristic of aromatic microcapsules for the functional textile applications have been reported by Zhao H. et al. [5]. It was found that the release rate of essential oil (Rose® 7289) from melamine-formaldehyde microcapsules significantly increased with the decrease in microcapsule particle size. As predicted according to the Peppas model, after one year of release, the small microcapsules with mean diameter of around 6.67 μm would be substantially depleted while larger microcapsules with mean diameter of around 37.69 μm would release only approximately 10% of the core material. Besides, the size of microcapsules also affected the textile impregnation efficiency and broken release intensity of microcapsule treated fabric. The last one was characterized by the peak height of 1,8-eucalyptin composition of essential oil (Rose® 7289) measured in the SPME-GC-MS elution curves. The composition 1,8-eucalyptin was considered as the essential oil quantity released from the microcapsule treated fabrics which had been followed the hitting process. The highest impregnation efficiency and broken release intensity were reached when the microcapsule size was similar to the textile fiber diameter (25 ÷ 30 μm) [5].

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Microcapsule size requirements in textile application depend on many factors, especially the use of end products and the structures of textile substrates. In general, microcapsules used in medical textile often have average diameters in range of 10 ÷ 60 μm [3], [4], [6]–[8]. Chu C. D. et al. [9] have proposed the suitable size of microcapsules, which was in the range of 15 ÷ 20 μm, for some popular knitted structures (Single, Rib1x1 and Interlock 1x1 fabrics). Their proposal based on the observation of the fabric structure by the optical microscopy and the scanning electron microscopy to determine the width and the height of the knitted loops.

Along with the size, the microcapsule morphology is also the interest of many studies about the microcapsule elaboration and application. Most of the models and equations used to predict and calculate the size and the release rate of active ingredients from microcapsules are based on an assumption that microcapsules are spherical in shape [10] [11]. However, for textile applications, the drying step, which is always obligatory in the finishing process, often makes microcapsules deflate significantly [4], [12]–[14]. Mollor P. et al. [13] studied the influence of drying temperature on the deformation of melamine formaldehyde microcapsules containing peppermint fragrance after being applied to cotton fabrics by padding technique. Three levels of drying temperatures investigated were 120, 140 and 160°C (all for 10 minutes). Their results showed that the presence of apparently unaltered microcapsules could be observed on the fabric treated by air at 120°C, some deflated microcapsules appeared when heated at 140°C and the swelling was reduced considerably in each microcapsule at drying temperature of 160°C. Based on FTIR spectrum of microcapsule padded fabrics at different drying temperatures, along with thermal analysis of microcapsules, the authors confirmed that microcapsule deformation was due to the fragrance loss from microcapsules during the drying step.

Microencapsulation by solvent evaporation method has been used to elaborate microcapsules for most of textile applications, including the field of medical textile products [7]. The basic principle of solvent evaporation technique from oil/water emulsion consists of two continuous stages: the emulsification stage and the solvent evaporation stage. In the first stage, an emulsion is created from the oil phase (a homogenous mixture of polymer, active ingredients, and a proper solvent) and the aqueous phase (a solution in water of a suitable surfactant). In the later stage, the emulsion is then subjected to reduced pressure and elevated temperature to gradually remove the solvent from the emulsion droplets. Eventually, when the droplet composition reaches the bimodal boundary, the polymer phase separates as small droplets of liquid, which are rich in solvent and polymer within the emulsion droplets. These droplets are mobile and migrate to the oil/water interface where they fuse and spread to engulf the original oil droplet if the wetting conditions are correct. Further solvent removal causes the polymer to precipitate at the interface, forming the shell [10].

Surfactant concentration and stirring rate are the two most important parameters that affect the size of microcapsules in the solvent evaporation method [10], [15], [16]. There is a general trend of many works indicating that the microcapsule size will decrease when the surfactant concentration increases because of the reduced interfacial tension. In the study of Valot P. et al. [15] on the microencapsulation of ibuprofen by solvent evaporation technique, the concentration of the PVA surfactant ranged from 0.1 to 2.0 wt%. Their results showed that the mean diameter of microcapsules was not influenced by the surfactant concentration above 0.2 wt%, whereas it significantly decreased below 0.2 wt%, the variation suggested that the droplet cover was insufficient at this level and could not prevent the droplet from coalescence. Besides, the PVA concentration also impacted the size distribution of microcapsules, which were relatively narrow only for the concentration of 0.2 wt%. In the study of Urbaniak T. and Musiał W. [16], the size of lavumidine-loaded microsphere was reported to decrease with the increase of homogenization speed. The microspheres were prepared by solvent evaporation method, using a laboratory rotor-stator homogenizer. When the homogenization speed increased from 10,000 to 35,000 rpm, the hydrodynamic diameter of microspheres decreased from around 2400 nm to 400 nm, respectively. This effect was explained by the increased shear stress at faster rotation rates, which caused the larger tangential stress, resulting in smaller emulsion droplets.

The halogenated solvents such as methylene chloride and chloroform have been the most used solvents for the solvent evaporation technique because of its high volatility, quite low boiling temperature and very low solubility in water. However, these solvents are confirmed carcinogenic according to EPA (Environmental Protection Agency) data and many researchers are making great efforts to find less toxic replacements [10]. Ethyl acetate is a non-halogenated solvent that has been used recently for microencapsulation by solvent evaporation method in the attempt to cut down the use of methylene chloride. Sah H. [17] was successful in producing PLGA microspheres by solvent evaporation method that used ethyl acetate as the solvent. The elaborated microspheres had the average diameter of 44 μm, which were obviously smaller than that of microspheres made by solvent extraction technique (93 μm). Moreover, the amount of residual solvent in the microspheres made by solvent evaporation technique, which was 2.62 wt%, was also much lower than that in the microspheres made by solvent extraction technique (6 wt%). Merabedini S. M. et al. [18] also made successfully

ethyl cellulose microcapsules containing plant oils by solvent evaporation method with the ethyl acetate solvent, the microcapsule size and the microencapsulation efficiency were equal to those of microcapsules made from the chloroform solvent. A big disadvantage of using ethyl acetate for the microencapsulation by solvent evaporation method is its high solubility in water (90 g/l at 20°C, which is 4.5 times higher than that of methylene chloride), which causes the fast diffusion of solvent from emulsion droplets to aqueous phase, leading to the fast precipitation of polymer wall, resulting in non-spherical microcapsules [17], [19]. The solvent diffusion rate could be slowed down by saturating the aqueous phase with a certain volume of ethyl acetate before the emulsification step [19], [20].

Quillaja saponin is a group of bio-sourced surfactants extracted from the *Quillaja Saponaria Molina* tree. With the presence of hydrophobic quillaic acid aglycone group and two hydrophilic sugar chains in molecular structure, quillaja saponin is amphiphilic and possesses surfactant properties due to the high surface activity and the ability to form micelles. Therefore, quillaja saponin has been used widely as stabilizer and emulsifier in the food and pharmaceutical fields [21]–[24]. However, so far, there has not been much research on the use of saponin as the surfactant in the microencapsulation.

This work aims to investigate the ability of quillaja saponin as natural surfactant and ethyl acetate as non-halogenated solvent for an eco-friendly microencapsulation by solvent evaporation method. The elaborated microcapsules of ibuprofen are oriented to medical textile applications due to their suitable size and morphology. The influence of saponin concentration, stirring speed during emulsification and volume of ethyl acetate added to aqueous phase before emulsification on the microcapsule size and morphology was investigated.

EXPERIMENTS

Materials

2-(4-Isobutylphenyl) propanoic acid (Ibuprofen) from BASF (Germany) and medium-chain triglyceride oil (Miglyol 812) supplied by SASOL (Germany) were used as model active ingredients, which help to diffuse the active agent ibuprofen and apply it to skin of user. The poly(ethyl acrylate-co-methyl methacrylate-co-trimethylammonioethyl methacrylate chloride) Eudragit RSPO polymer was from EVONIK Industry (Germany). Quillaja saponin ($C_{36}H_{54}O_{11}$) from *Quillajar* bark (Sapogenin content 20 ÷ 35 %) was purchased from SIGMA-ALDRICH. Ethyl acetate solvent with purity of 99.9 % was supplied by CARLO ERBA. All chemicals for the microencapsulation process have been used as providing without any more purification.

The textile substrate used was cotton interlock fabric knitted from cotton yarn (Ne20) with the loop length of 3.78 mm, the horizontal density of 122 wales/10 cm and the vertical density of 147 courses/10 cm. The interlock fabric meter square weight was 396 g/m2. The knitting process were carried out on the flat knitting machine SSR-112 (knitting gauge R16) of Shima Seiki (Japan). The grey fabrics were then scoured and bleached at Doximex Knitting Company (Vietnam).

Microencapsulation

The microencapsulation based on the solvent evaporation method, which was often used to elaborate the ibuprofen-loaded microcapsules. Ethyl acetate solution (15 ml) containing ibuprofen (8.33 mg/ml), miglyol 812 (33.33 mg/ml) and eudragit RSPO (116.67 mg/ml) was added dropwise for 5 minutes to 100 ml of a quillaja saponin aqueous solution under blade stirring at 700 rpm. The evaporation of ethyl acetate at reduced pressure (300 ÷ 350 Torr) was initiated 5 minutes after the emulsification start with a stirring rate of 600 rpm. Microcapsules collected after 5 hours of evaporation were washed three times with distilled water and then preserved in type of microcapsule suspension in the lab fridge.

To investigate the influence of quillaja saponin concentration on the characteristics of microcapsules, saponin concentration varied by four levels of 0.025, 0.05, 0.075 and 0.1 wt%, the microcapsule lots were coded as C0.025, C0.050, C0.075 and C0.100, respectively.

To investigate the influence of stirring rate during the emulsification stage on microcapsule characteristics, the stirring rate changed in three levels of 700, 650 and 600 rpm, the correlative microcapsule lots were coded as R600, R650 and R700.

To investigate the influence of the solvent volume added to the continuous phase before emulsification on microcapsule characteristics, the volume of ethyl acetate added to the saponin solution was manipulated by 0 ml, 8 ml and 12 ml, and the correlative microcapsule lots were coded as S0, S8 and S12.

Application of microcapsules to the fabric

The washed microcapsules were re-dispersed in distilled water to make the microcapsule suspension with concentration of 12.5 mg/ml. Circle fabric sample with diameter of 25 mm was placed into a plastic cup having the same diameter. The microcapsule suspension (5 ml) was poured into the cup. After 12 hours of soaking, the fabric sample was taken out and then dried in a vacuum drier at 25°C until totally dry.

Microcapsule characterization

The microcapsule suspension was diluted two times for the optical microscopy observation with an

Olympus EX microscope. The microcapsule morphology was also observed by scanning electron microscopy on an ESEM (XL SERIES – Philips) at low vacuum mode (3 ~ 3.5 Torr, 15 kV).

The microcapsule - treated fabric was observed by Olympus EX microscope and by the scanning electron microscope (SEM) QUANTA FEG 250.

The average diameter and the size distribution of microcapsules were determined by static laser light scattering using a Mastersizer 2000 (Malvern instruments, UK). The broadness of the size distribution curve was expressed by Span value calculated as below:

$$Span = \frac{d(0.9) - d(0.1)}{d(0.5)} \quad (1)$$

In which, *d* (0.5) indicated that 50% of total particles were smaller than this size, it was similar for *d*(0.9) and *d* (0.1) value.

According to (1), the smaller span value represented the narrower size distribution, meaning that the size of microcapsules was more homogeneous.

RESULTS AND DISCUSSION

Influence of saponin concentration on the microcapsule size and their morphology

The surfactant concentration has been reported to have strong effect on the size of emulsion droplets and, consequently, the size of elaborated microcapsules [10], [15], [16]. For this work, in order to reveal the influence of saponin concentration on the microcapsule size and morphology, the saponin concentration varied by four levels of 0.025, 0.050, 0.075 and 0.100 wt%. The microcapsule lots were coded by C0.025, C0.050, C0.075 and C0.100, respectively.

The *d*(0.5) diameter and the span values of four microcapsule lots were presented at Table 1 and the influence of saponin concentration on the microcapsule size distribution was shown in Fig. 1.

It could be seen that the average diameter of elaborated microcapsules was in range of 17.1 ÷ 34.3 µm, which was popular for most medical textile applications [3]–[5]. When the saponin concentration increased from 0.025 to 0.1 wt%, the *d*(0.5) diameter of microcapsules decreased respectively from 34.3 to 17.1 µm. This result was similar to the trend reported in some literatures for various types of surfactant, including quillaja saponin [15], [16], [25]–[27]. At high concentration, more surfactant molecules could be oriented at the interface of aqueous - oil phases to reduce efficiently the interfacial tension, which resulted in the formation of smaller emulsion droplets, and then smaller microcapsules.

Table 1. Effect of saponin concentration on the diameter and size distribution of microcapsules.

Lot	Saponin concentration [wt%]	*d*(0.5) diameter [µm]	Span value
C0.025	0.025	34.3 ± 0.3	2.4
C0.500	0.050	23.2 ± 0.2	2.8
C0.075	0.075	21.5 ± 0.2	3.0
C0.100	0.100	17.1 ± 0.2	3.9

The decrease of the microcapsule *d*(0.5) diameter was especially important when the saponin concentration increased from 0.025 to 0.05 wt%. The significant size decrease (from 34.3 to 23.2 µm) might be connected to the cmc value of quillaja saponin from Sigma Aldrich, which was reported about to 0.05 wt% [28]. With a saponin concentration lower than the cmc value, the droplet surface was only partially covered, the interfacial tension between the organic and aqueous phases decreased significantly according to the addition of surfactant concentration. When the saponin concentration reached the cmc value, the effect of the increase in saponin concentration became less significant on the interfacial tension, and consequently, did not quite affect the microcapsule size.

The optical microscope images (Fig. 2) helped to confirm the results of laser diffractometry. Besides, the optical microscope images revealed that most of microcapsules were in spherical shape while some of them were in elliptical or rod shape. Since most equations used to predict the active release rate from microcapsules were only based on spherical ones, those irregular microparticles would be a big disadvantage in predicting the drug release profile from microcapsule-treated fabric.

The similar phenomenon was mentioned in some referred researches on the microencapsulation by solvent evaporation/extraction methods with the use of ethyl acetate [17], [19].

The solubility in water of ethyl acetate (90 g/l at 20°C) was much higher than that of halogenated solvents most used in solvent evaporation method such as dichloromethane (20 g/l at 20°C). With high stirring rate during the emulsification, ethyl acetate in oil droplets diffused quickly to the aqueous phase, inducing the fast polymer precipitation and eventually, increasing the formation of elliptical or rod shape microparticles as shown in Fig. 2.

Span value was quite high in the range of 2.4÷3.9 (Table 1), which presented the broad size distribution of microcapsule lots. The fast polymer precipitation could be a reason for this phenomenon because the emulsion droplets did not have enough time to be divided into smaller and more stable ones.

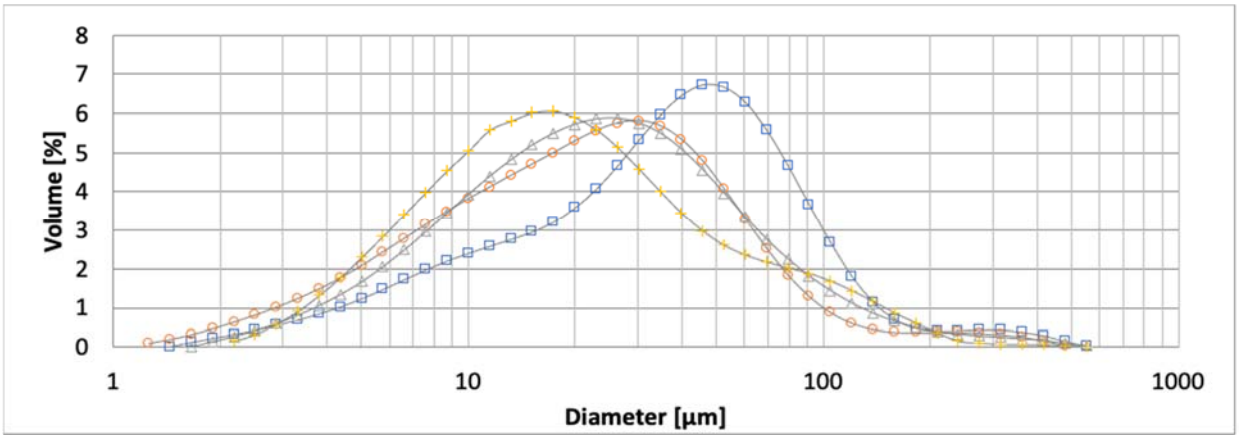
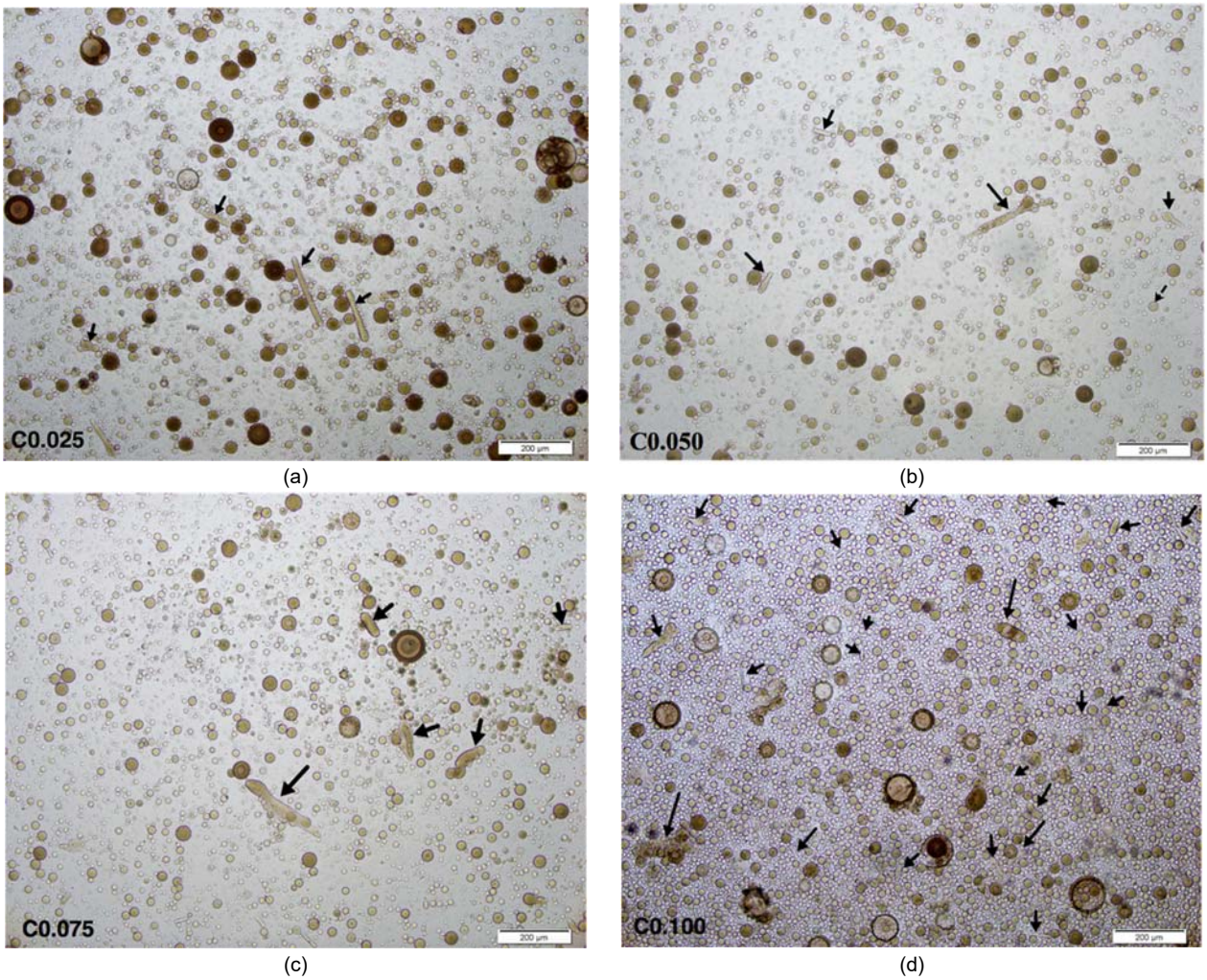


Figure 1. Size distributions of microcapsules at different saponin concentrations.



(a) (b) (c) (d)

Figure 2. Optical microscope images of microcapsules obtained at different saponin concentrations. C0.025 (a), C0.050 (b), C0.075 (c) and C0.100 (d). ⟶: irregular microparticles.

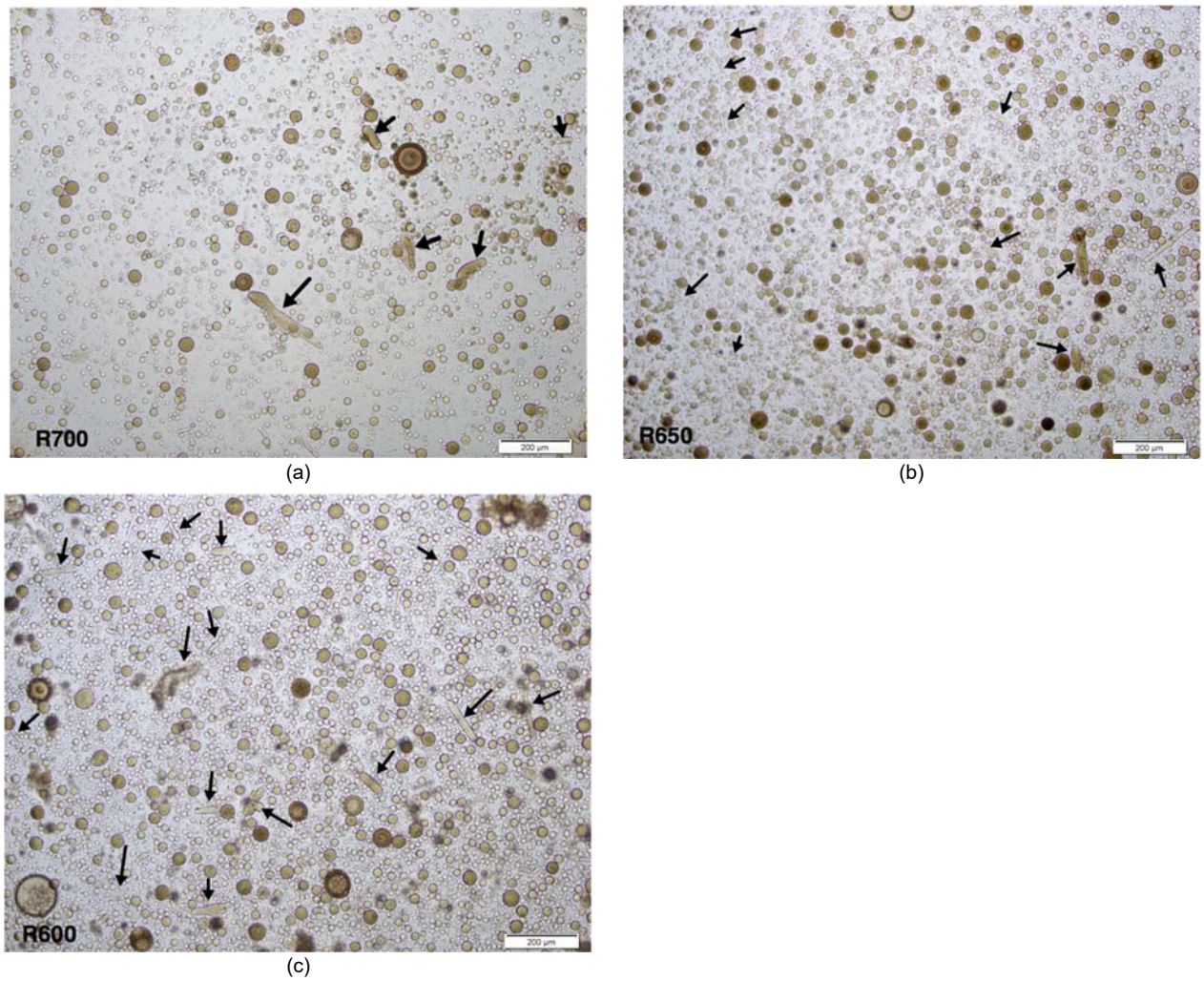


Figure 3. Optical microscope images of microcapsules obtained at different stirring rates. R700 (a), R650 (b) and R600 (c). ⟶: irregular microparticles.

Moreover, the span value increased at higher concentration of saponin surfactant and the most incline was from 3.0 to 3.9 when saponin concentration varied from 0.075 to 0.1 wt%. Might be the high saponin concentration above the cmc value (0.05 wt%) promoted the solubility of ethyl acetate in water, accelerating the diffusion of solvent from oil droplets to the aqueous phase and the polymer precipitation, resulting in broader microcapsule size distribution. This assumption will be verified in a further research.

Influence of stirring rate during emulsification stage on microcapsule size and morphology

As discussed above, high stirring rate during the emulsification step could be a reason for the presence of irregular microparticles in microcapsule lots. The high shear stress induced by the strong stirring action not only accelerated the diffusion of ethyl acetate solvent from the oil droplets to aqueous phase but also pulled along the oil droplets, resulting in the formation of irregular microparticles [19]. So, in order to reduce the number of the non-spherical microparticles and to get a narrower size distribution, the influence of stirring rate on microcapsule size and morphology was investigated. Because the rate of 700 rpm was nearly the upper limit of the mechanical stirrer, three levels of stirring rate investigated were: 700, 650 and 600 rpm. The correlative microcapsule lots were coded as R700, R650 and R600, with a saponin concentration equal to 0.075 wt%.

The optical microscope images of three investigated microcapsule lots were shown in Fig. 3 and it could be seen that reducing stirring rate did not help to decrease the number of irregular microparticles as expected.

Besides, reducing stirring rate from 700 to 600 rpm did not seem to affect the mean diameter of microcapsules, which was still about 21 ÷ 22 µm (Table 2). However, the size distribution became broader (Fig. 4) with the span value increased considerably from 3.0 to 3.9 (Table 2), it could be due to the less uniform mixing force throughout the emulsion mixture at low stirring speed.

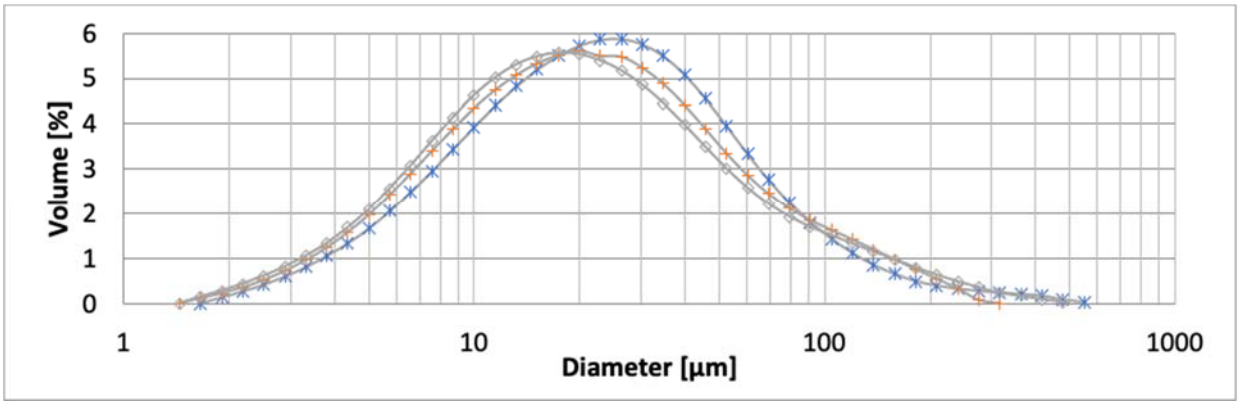


Figure 4. Effect of stirring rate on size distributions of microcapsules.

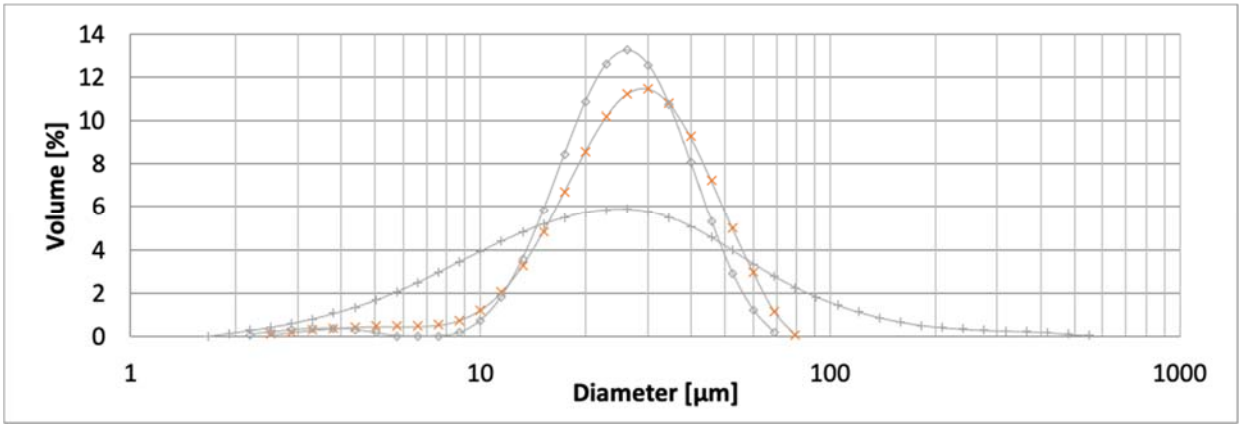


Figure 5. Effect of stirring rate on size distributions of microcapsules.

Table 2. Effect of stirring rate on the diameter and size distribution of microcapsules.

Lot	Stirring rate during emulsification [rpm]	*d*(0.5) diameter [µm]	Span value
R700	700	21.5 ± 0.2	3.0
R650	650	22.4 ± 0.2	3.5
R600	600	21.0 ± 0.1	3.9

Influence of the volume of ethyl acetate added to the aqueous phase

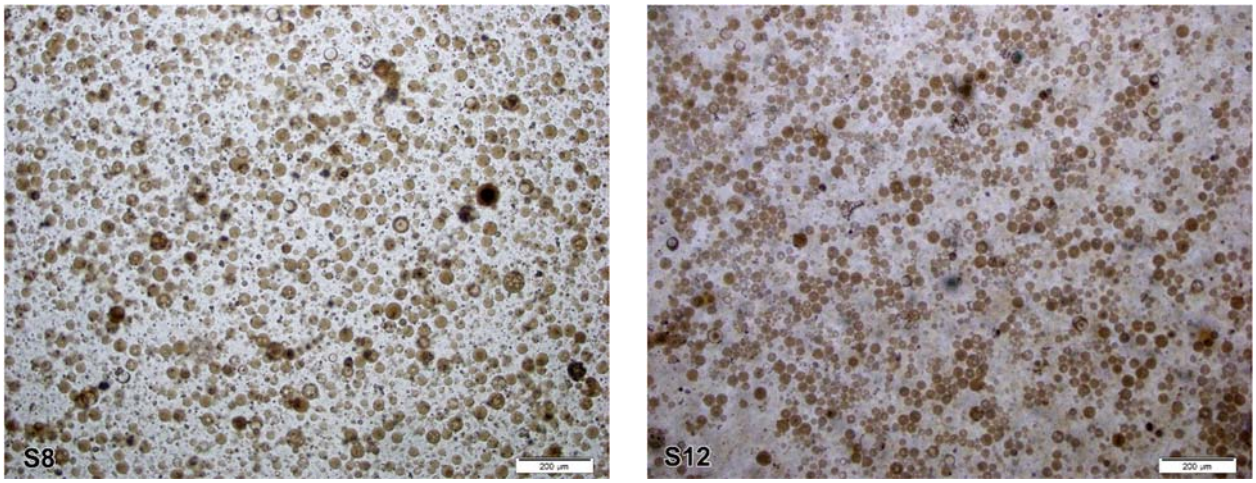
The fast diffusion of ethyl acetate from oil droplets to aqueous phase was the main reason for the formation of elliptical and rod shape microparticles because it induced the rapid polymer precipitation, so disturbing the emulsification step. In literature, the diffusion rate was reported to be slowed down by saturating the outer aqueous phase with a certain volume ethyl acetate prior to the emulsification step [19], [20]. Therefore, in this work, a certain volume of ethyl acetate was added to the aqueous phase before the emulsification step for an effort of reducing the irregular microparticles and narrowing the size distribution. At saponin concentration of 0.075 wt% and the stirring rate of 700 rpm, the volume of ethyl acetate added to the aqueous phase varied by three levels: 0 ml (without saturation of aqueous phase with ethyl acetate), 8 ml and 12 ml. The microcapsule lots were coded by S0, S8 and S12, respectively. It should be noted that the S0 microcapsules were exactly the C0.025 and R700 microcapsules in the previous investigation.

Table 3. Effect of ethyl acetate volume on the diameter and size distribution of the microcapsules.

Lot	Volume of ethyl acetate added to the aqueous phase [ml]	*d*(0.5) diameter [µm]	Span value
S0	0	21.5 ± 0.2	3.0
S8	8	29.5 ± 0.2	1.3
S12	12	27.5 ± 0.2	1.1

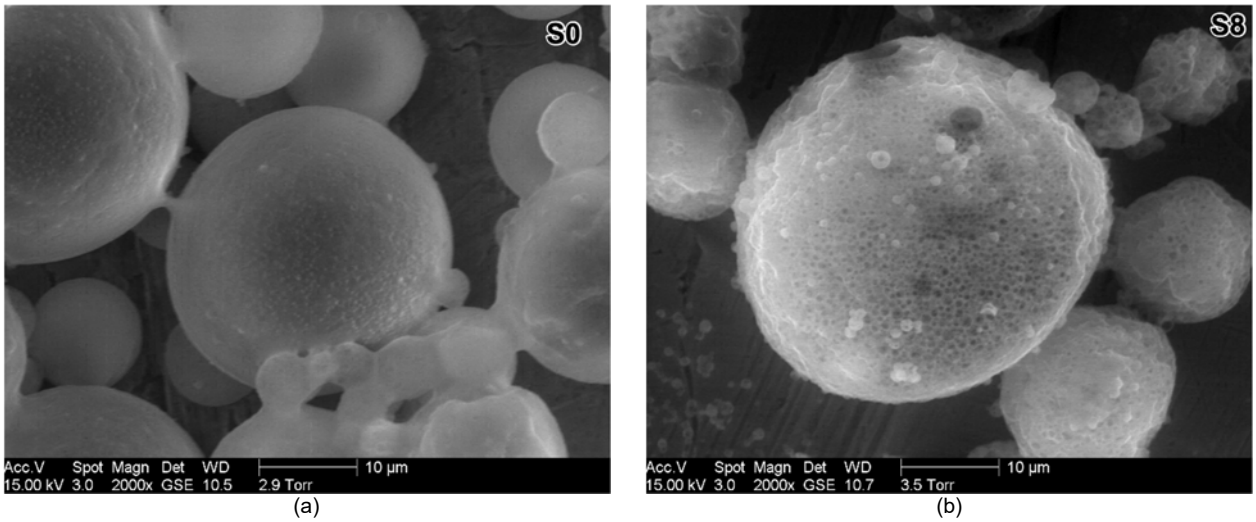
The mean diameter and size distribution of microcapsule lots were presented in Table 3 and Figure 5.

According to data in Table 3 and Fig. 5, the addition of ethyl acetate to the aqueous phase before emulsification really helped to narrow the microcapsule size distribution. When 8 ml of ethyl acetate was added to the aqueous phase, the span value decreased obviously from 3.0 (for S0 microcapsules) to 1.3 (for S8 microcapsules) while the *d*(0.5) diameter of microcapsules increased slightly from 21.5 µm (for S0 microcapsules) to 29.5 µm (for S8 microcapsules). However, with more ethyl acetate added to the aqueous phase (12 ml for S12 microcapsules), the microcapsule diameter (27.5 µm) and the span value (1.1) did not change much.



(a) (b)

Figure 6. Optical microscope images of microcapsules S8 (a) and S12 (b).



(a) (b)

Figure 7. SEM images of microcapsules S0 (a) and S8 (b).

The optical microscope images and S8 and S12 microcapsules (Fig. 6) showed the similar trend obtained by the laser diffractometry. Moreover, it was revealed that the irregular microparticles almost disappeared in the S8 and S12 microcapsule lots.

Since the solubility in water of ethyl acetate at 20°C is 90 g/l that equals to 100 (ml/l), with 8 ml of ethyl acetate added to 100 ml of aqueous phase, the solvent was completely dissolved and almost reached the water saturation. The diffusion rate of ethyl acetate from the oil droplets to the aqueous phase was then significantly reduced, resulting in the delay of the polymer precipitation, increasing the efficiency of the emulsification step through the formation of smaller droplets with spherical shape. With more than 8 ml of ethyl acetate was added to the aqueous phase (for example in case of S12 microcapsules), the aqueous phase might be completely saturated by the solvent, so the mean diameter and the size distribution of microcapsules did not change much in comparison to the S8 lot.

SEM images of S0 microcapsule (Fig. 7(a)) and of S8 microcapsule (Fig. 7(b)) showed the clear difference in the microcapsule surface morphology.

The external surface of S0 microcapsules (without saturation) appeared quite smooth (Fig. 7(a)) while S8 microcapsules (saturating the aqueous phase by 8 ml of ethyl acetate) exhibited very porous surface with many pinholes larger than 1 µm (Fig. 7(b)). This difference also was reported in literatures [29], [30] in which, the fast removal of solvent from oil droplets tended to form the smoother surface of microcapsules and in contrast, the slow removal of solvent would cause the microcapsule structure more porous. Besides, in the case of ethyl acetate, the high miscibility between ethyl acetate and water favored the uptake of water into the dispersed phase and created water pockets inside the microcapsule core when the polymer precipitated. The more water entrapped inside the droplets, the more porous the microcapsule would be. The saturation step delayed the solvent removal during the emulsification step

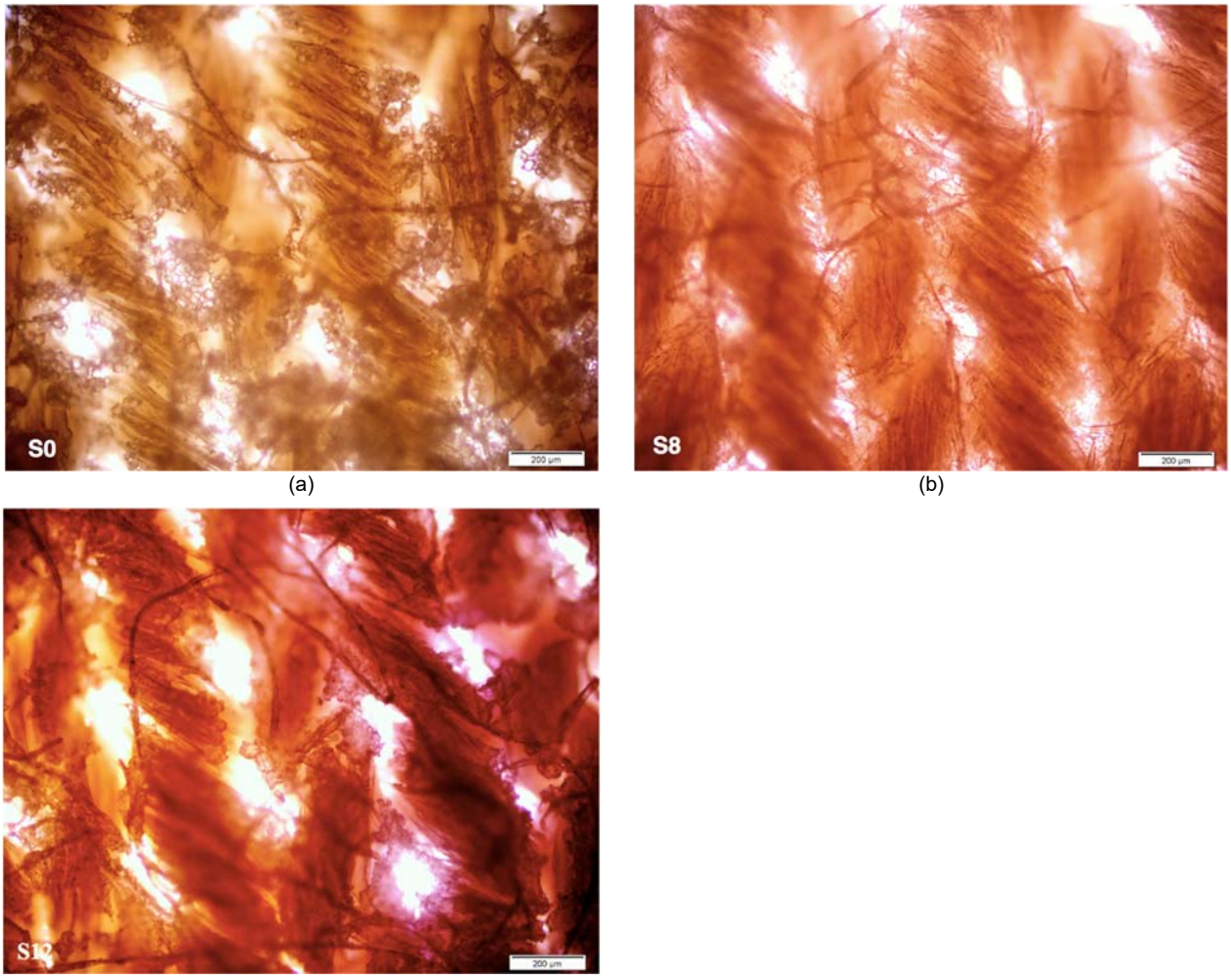


Figure 8. Optical microscope images of cotton interlock knitted fabric treated with microcapsules obtained at different ethyl acetate added volume.

and therefore slowed down the solidification of droplets, favoring the water diffusion into the semisolid microcapsules that eventually created more porous microcapsules.

The S0, S8 and S12 microcapsules were then applied to cotton interlock knitted fabric to test their drying ability during the fabric finishing process. The microscopic images of microcapsule treated fabrics were presented at Fig. 8.

It was shown that after the fabric treatment, which required a vacuum drying stage at 25ºC for 24 hours, the S0 microcapsules were still spherical (Fig. 8(a)) while the S8 and S12 microcapsules deformed strongly, no completely spherical microcapsules could be observed on the fabric surface (Fig. 8(b,c). The addition of ethyl acetate to the aqueous phase decreased the diffusion rate of solvent from the oil droplets to the aqueous phase. After the same duration of solvent evaporation step (5 hours), the content of residual solvent in S8 and S12 microcapsules were higher than that in S0 microcapsules. Since the water solubility in ethyl acetate is equal to 3.3 wt% [17], the content of residual water in S8 and S12 microcapsules might also be higher than that of S0 microcapsules. Therefore, the polymer structure of S8 and S12 microcapsules was softer and weaker than of S0 microcapsules, resulting in their serious deformation after the drying process.

CONCLUSION

In conclusions, it was successful to produce microcapsules by solvent evaporation method using quillaja saponin as bio-sourced surfactant in the combination with ethyl acetate as non-halogenated and less toxic solvent. The microcapsules were in spherical shape with the size range of 17 ÷ 34 µm, suitable for the medical textile applications.

When the saponin concentration increased from 0.025 to 0.1 wt%, the $d(0.5)$ diameter of microcapsules decreased from 34.3 to 17.1 µm while the span value increased from 2.4 to 3.9, respectively. The irregular microparticles with elliptical or rod shapes were observed in all four microcapsule lots.

The stirring rate during emulsification step was slowed down gradually by 700, 650 and 600 rpm in an effort of reducing the number of irregular microparticles, but it did not work. The *d*(0.5) diameter almost did not change at around 21.5 μm while the span value increased quickly from 3.0 to 3.9.

Another solution to overcome the formation of irregular microparticles was saturating the aqueous phase by a certain volume of ethyl acetate prior to the emulsification step. With 8 and 12 ml of ethyl acetate added to the aqueous phase, the irregular microparticles almost disappeared in the microcapsule lots. The *d*(0.5) diameter increased slightly from 21 to 28 μm while the size distribution of microcapsules became obviously narrower with span value decreased from 3.0 to 1.1. However, the saturation step made the microcapsules more porous, softer, and weaker, so they were easily deformed during the drying stage, leaving a polymer coating on the fabric surface rather than individual dried microcapsules after the fabric treatment.

For the medical textile applications, the microcapsules always need to pass a drying process. Therefore, in the scope of research, despite of the broad size distribution and the formation of some irregular microparticles, the microcapsule lot C0.075 (R700 or S0) was chosen to be the best candidate for medical textile applications due to their capability of keeping spherical shape during the drying step.

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