Microencapsulation of active substances and fragrances in textile material applications

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Microencapsulation of substances applied on textiles is a relatively new method with many advantages applied in many different fields. There are many effective approaches to microencapsulation for decreasing release of fragrances and active substances such as essential oils and cosmetic ingredients using cyclodextrins, yeast cells, chitosans, melamine-formaldehyde or poly(L-lactide) as wall materials. Some of the methods for encapsulating are: complex coacervation, phase separation, interfacial polymerization and in situ polymerization, spray drying, spray congealing and pan coating. Market products for textiles with microcapsules include cosmetotextiles, aromatherapy textiles, home textiles, sports wears and apparel and microcapsules can be applied to these textiles by padding, coating, spraying or immersion exhaustion techniques. In practice the aim is to produce textiles with microcapsules which would last for as much wash and dry cycles as possible.
Key words: *microencapsulation, active substances, fragrances, textile applications*

1. Introduction

Microencapsulation is a process of packing small droplets of liquids or solid particles into a thin film which may be made with various coatings. The outer part of a microcapsule is called a shell or a wall, and the filling is called a core (Fig.1). The choice of the wall material depends on the core material, its release method and the use of the product [1, 2].

Microcapsules are used in many different fields, e.g.: pharmaceuticals, cosmetics, agrochemicals, and flavors and essences. Some types of microcapsules are put in food, other in cosmetics e.g. in shower gels, and yet others are applied on fabrics. Some of the most common substances put into microcapsules in materials are skin softeners, repellents, antimicrobial agents, dyes, vitamins, hormones and other drugs, as well as fragrances [2, 3]. However, material applications include not only textiles, but also paper, band aids etc.

Adding fragrances to textiles was introduced a long time ago, but fragrances remained present only for one or two wash cycles. Application of microcapsules with fragrances allowed to extend the life time of the product to even 30 wash cycles and to three to five years if the product is on the shelf [4]. The majority of the work has been in microencapsulated 'scratch and sniff' T-shirts and in women's hosiery. Many companies applied microcapsules with fragrances to sofas, curtains, cushions, sheets and some toys. In the early days the applications included drawer liners,

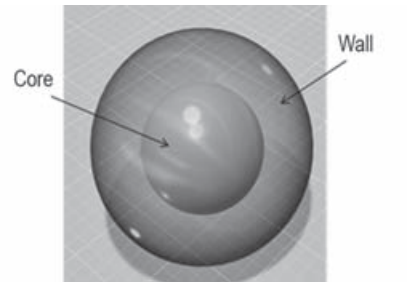

Fig.1 Structure of a microcapsule

paper handkerchiefs, gift wrapping, stationary, greeting cards, advertising brochures, books, cartons and labels [3].

Marketed product for textiles with microcapsules include among others cosmetotextiles, aromatherapy textiles, home textiles, sports wears and apparel [5]. In the first group there are microencapsulated skin moisturizers, vitamins, provitamins and antiaging substances. The purpose of these textiles is to contact with the skin. In other groups there are textiles with microencapsulated fragrances or, as in the case of home textiles, also deodorizing substances [6].

This article is a review of microencapsulating active substances, used in applying on materials, especially textiles.

2. Composition of microcapsules

There are many effective methods of microencapsulation which postpone the release of fragrances, but cyclodextrins (CD) are the best regarding safety of the human body, because β-cyclodextrin does not cause skin irritation, skin sensibilisation or mutagenic effect [7]. The possibilities of application to the textile industries using cyclodextrins has become more interesting [8].

Wang and Chen worked on textiles with aromatherapeutic properties. In their research, the fragrance with β-cyclodextrin inclusion compounds were fixed onto cotton with a low-temperature binder by the conventional padthermofixed method. That binder was a polymer. Scent release was measured every 5 days for a month. Even though the scent intensity decreased, the release of fragrances lasted over 30 days [7].

Lee and his coworkers embedded β-cyclodextrins as acrylamidomethyl CD onto cellulose fibers using a chemical bonding instead of physical attachment. Benzoic acid with its antimicrobial activity and vanillin as the aroma agent were encapsulated. The antibacterial activity investigated in the presence of *Escherichia coli* and *Staphylococcus aureus* was retained after 10 wash cycles. The fragrance of the encapsulated vanillin remained present during storing for 7 day at room temperature and the next 7 days at the temperature of 80 °C [9].

The idea of microcapsules whose walls were made with chitosans was patented by Copete Vidal et al. Membranes were made with the use of salts of alginic acids. Microcapsules containing various active components were embedded onto the fibers with a binder. It was examined that using a binder to embed microcapsules reduces the degree of releasing active substances from the fiber both in machine and hand washing [10].

Nelson used waste yeast cells of *Saccharomyces cerevisiae* in microencapsulation in textiles. After encapsulating the core material, yeast cells were attached to both cotton and wool fibers by using crosslinking agents and binders [3, 11].

Hong and Park prepared me-lamine-formaldehyde as wall materials with Migrin oil as a core and applied it on cotton fabrics. Microcapsules were prepared by in situ polymerization. One of the examined parameters of capsules with fragrant oil was the size of particles which remain 10 μm even after 15 wash cycles [12].

Hong and Park investigated also poly(L-lactide) microcapsules with Forest-shower fragrance. They were pre-pared by the interfacial precipitation method through solvent evaporation from (w/o)/w emulsion and embedded on cotton fabric. The sizes of most particles were below 5 μm and remained the same even after 15 cycles of washing [13].

3. Microencapsulation methods

An important part of microencapsulating of essential oils and fragrances is how to contain all of ingredients in the capsules [6].

Some of the microencapsulation methods are: complex coacervation, phase separation, interfacial polymerization and in situ polymerization, spray drying, spray congealing and pan coating [11, 14, 15].

3.1. Complex coacervation

Complex coacervation is a method of microencapsulation in which a core is completely surrounded by a continuous coating of a wall. It allows to encapsulate up to 99 % of active substances inside the wall [11, 16, 17].

This method is mainly used to microencapsulate substances with hydrophobic properties. Complex coacervation is based on the interaction between different polymers with opposite charges. This interaction forms insoluble complexes and phase separation, and relies among other things on the charges carried by the biopolymers, the ratio between them, the pH of the coacervation mixture, the ionic strength, and the accessibility of the charges for the interaction. The deposition of such complexes around a hydrophobic core creates a barrier, which enables the encapsulation. The biopolymers which are used in the coacervation process have hydrophilic colloidal properties, solubility in aqueous medium, adequate charge density and linear chains [17, 18].

Microcapsules produced in the coacervation process have excellent controlled release characteristics and heat resistant properties [19]. Comparing to other microencapsulation technologies such as spray drying, coacervation is a gentle process because no high heat is involved. Because of that, this process may be used to encapsulate flavor oils but also fish oils, nutrients, vitamins, preservatives, enzymes etc. [17].

3.2. Phase separation

Phase separation is a physicohemical process of making microcapsules. It allows to produce particles of 2-5000 μm size and with liquid core [15].
Two polymers which are not mixing with each other are dissolved in a common solvent. These polymers form two distinct liquid phases. One phase is rich in polymer and designed to act as the capsule shell, while the other is rich in incompatible polymer. The incompatible polymer is presented in the system to cause the formation of two phases. It is not designed to be part of the final capsule shell, although a small amount may remain entrapped in the final capsule as an impurity. The process is normally carried out in organic solvents and used to encapsulate solids with a finite degree of water solubility [11].

3.3. Interfacial Polymerisation

In the interfacial polymerization the shell is formed from multifunctional monomers which are dissolved in the liquid core material and then dispersed in an aqueous phase with a dispersing agent. A rapid polymerization reaction is then produced at the interface which finally generates the capsule shell. Although this method allows to encapsulate both the liquid and solid, the chemistry of the polymerization is different [11, 15, 20]. This method is widespread in industrial use, but it cannot be used to encapsulate sensitive actives [20].

3.4. In Situ Polymerisation

Similar to interfacial polymerization, the in situ polymerization involves preparing the wall of a capsule through polymerization of monomers which are added to the encapsulation reactor. However, reactive agents are not added to the core material [11, 15].
Polymerization occurs exclusively in the continuous phase side of the interface formed by the dispersed core material and continuous phases. A low weight prepolymer is produced in a polymerization process and during growing in size it deposits onto the surface of the dispersed core material being encapsulated, where polymerization with crosslinking continues to occur, thereby generating a solid capsule shell [11, 15].

3.5. Spray drying

Spray drying is the physicomechanical method commonly used for microencapsulation where an active material is dissolved or suspended in a melt or polymer solution and becomes trapped in the dried particle [11, 21]. Particles produced in this process are the size 1-150 μm [21].
In this process an emulsion is created from the liquid product to be treated, a carrier substance and a filmogen solution. This emulsion is sprayed into small droplets in hot air. In high temperature, the solvent evaporates leading to a solid matrix around the dispersed second phase. It causes the small droplets of the product to be stored in the carrier substance and embedded in the filmogen [22].
The main parameters of the process are the core and the wall material, the temperatures of drying air of inlet and outlet, the gas flow rate, the distribution of temperature and humidity inside the dryer, the time of residence, and the geometry of the drying chamber [21].
It is desired for the wall material to have good emulsifying and film-forming properties, be easy to dry with low viscosity at high solid concentrations and chemically inert [21].
An advantage of this process is the short time of contact time in the drier, which allows to use this method to encapsulate labile materials. It is possible to use nitrogen gas as the hot gas instead of air if the encapsulated substances are sensitive to oxygen. However, some of the low-boiling point aromatic substances can be destroyed in the spray drying process [11, 23].

3.6. Spray congealing

Spray congealing consists of atomization of a fluid into an environment maintained at a temperature below the carrier melting point. The atomization leads to the formation of molten droplets which then solidify upon cooling, producing the final microparticles [14].

3.7. Pan coating

Pan coating is widespread in the pharmaceutical industry. With this method tablets, capsules, multiparticulates and drug crystals are produced [11].
Coating is a process in which a coating solution is applied to a solid core material (e.g. tablets) in a coating pan and the tablet surfaces become covered with a polymeric film. Before the tablet surface dries, the applied coating changes from a sticky liquid to tacky semisolid, and eventually to a nonsticky surface. The entire coating process is conducted in a series of mechanically operated coating pans. The smaller pans are used for experimental, developmental, and pilot plant operations and the larger once are used for industrial production [15].

4. Application in textiles and issues related to them

Microcapsules can be applied to textiles by padding, coating, spraying, immersion or exhaustion. A binder is required for all these methods. The binder may be acrylic, polyurethane, silicone, starch, etc. Its role is to fix the capsules onto the fabric and to hold them in place during wear and washing. Microcapsules can be applied to various fibers both natural and synthetic [3, 4].
Release mechanisms of the core for cosmetic textiles are friction, pressure, biodegradation and for aromatherapy and fragrance textiles these methods are friction and diffusion through polymer wall [11].
PCM (phase change materials) and antimicrobial materials were produced using late injection of microcapsules into the fiber and loading it with 5-10 % of microcapsules. This allowed to eliminate such processes

as knitting, spinning or dyeing. Using this method did not change normal properties of the fiber (strength, softness and drape) [24].

The smaller the microcapsules, the greater the covering of the product and the longer the fragrance will last, as it takes longer for the capsules to be ruptured by physical pressure [3]. In practice the aim is to produce textiles with microcapsules which would last for as many wash cycles as possible.

The Matsui Shikiso Chemi-cal Co of Kyoto has developed a way of fixing microcapsules with aroma to a fabric. The fabric is first treated with a nitrogenous cationic compound and the microcapsule wall is manufactured to adhere to this layer. The produced capsules are 0.1 to 100 μm in size and are made using interfacial or in situ polymerization methods. Typical encapsulated compounds include perfumes [3].

Conventional fixation during finishing the fabric is associated with high temperatures, which may destroy the fragrant. Li and coworkers prepared cotton fabric with encapsulated lemon fragrant on it and fixed it with an UV resin in the presence of initiator. UV resin may be cured in low temperatures. Scientists investigated that using that resin with an initiator allows the fragrant to remain on the fabric for more than 50 wash cycles compared to 25 cycles of washing for thermal curing of a fiber [25].

The market products of microencapsulating in textiles that are the topic of this article include cosmetotextiles, aromatherapy textiles, home textiles, sports wears and apparel [5]. In the first group there are included microencapsulated skin moisturizers, vitamins, provitamins and anti-aging substances. A purpose of this textiles is to contact with skin. In other groups there are textiles with microencapsulated fragrances or as in the case of home textiles also deodorizing substances [6].

In the market there are many companies which produce scented and cosmetic textiles, both natural and synthetic for clothing, undergarments, furnishings, bedding, accessories, etc. Microcapsules with active ingredients or essential oils have polymer walls and their diameters are about 2 μm. They are put into textiles using padding, immersion, spraying or recharging. The cores of microcapsules are released during wearing due to their rupture. Cosmetic textiles line includes aromatherapy for leg comfort, energizing, relaxing and respiratory treatment [26].

Some companies prints cosmetics catalogues with scented paper to smell perfumes. To print a page like that it is needed to use a scented lacquer in which there are microcapsules with an aroma oils as cores. After rubbing the page, walls of microcapsules rupture and the scent is remained [27].

There are many more companies that produce fibers with microcapsules with fragrances or cosmetics. They produce such products as scarfs with perfumes, perfumed dresses, bras and moisturizing and energizing tights and also underwear with microcapsules with silk to reduce cellulite and smooth the skin [3, 28].

As it was mentioned not only fragrances, essential oils and cosmetic compounds may be encapsulated. Also hormones and vitamins may be a core of a microcapsule. It would be good to apply medicines on textiles and wear them when we need it and not being worried of taking pills with those medicines which may cause destroying the liver or stomach.

However, there are also some problems related to microencapsulation compounds.

The most common issue is how to increase the durability of capsules during washing and drying. Nowadays, according to researches, these products withstand only for about 30 wash cycles. Increasing the durability involves the proper choice of wall material and the methods of preparing and applying microcapsules on materials. This areas of microencapsulating are constantly being developed.

Taking into account microencapsulated fragrances and essential oils there is a risk of allergies in contact them with the human skin. These substances, mainly essential oils which are natural products may contain some of 26 potential fragrance allergens. These substances may cause among others skin irritations, rash or swelling.

5. Conclusion

Microencapsulation of substances applied on textiles is a relatively new method with many advantages. It allows to create innovative products with new properties. Hoever, there are also some problems related to this method.

Despite problems of allergies and durability of microcapsules applied on textiles it is still an innovative method which may influence many areas of life and may cause development of the cosmetic and textile sector. It may also develop the pharmaceutical and medicine sector if textiles with microencapsulated medicines will be widely accessible for users.

References:

[1] Jyothi SriS. et al.: Microencapsulation: A Review. International Journal of Pharma and Bio Sciences 3 (2012) 1, 510-531

[2] Hammad U. et al.: Microencapsulation: Process, Techniques and Applications. Internatio-nal Journal of Research in Pharmaceutical and Biomedical Sciences 2011, 2 (2): 474-481

[3] Nelson G.: Application of microencapsulation in textiles. International Journal of Pharmaceutics (2002) 242, 55-62

[4] Ocepek B., P. Forte-Tavčer: Microencapsulation in textiles. XVIth International Conference on Bioencapsulation 2008, Ireland: 1-4

[5] Desai AA.: Medical Textiles: Healthcare And Hygiene Products. www.fibre2fashion.com, December (2013)

[6] Bolenwar P. et al.: Fragrance finishing of textiles. www.fibre2fashion.com, November (2013)
[7] Wang CX., ShL. Chen: Aromachology and its Application in the Textile Field, Fibres & Tetiles in Eastern Europe January (2005) 13, 6 (54): 41-44
[8] Shahba AF.: Production and Characterization of Novel Perfumed Curtain Fabrics. RJTA 12 (2008) 4, 31-40
[9] Lee MH. et al.: Grafting onto Cotton Fiber with Acrylamidomethylated -Cyclodextrin and Its Application. Journal of Applied Polymer Science (2000) 78, 1986-1991
[10] Vidal C. et al.: Equipped fibers and textile surface structures. Patent 0150056 A1, USA 2005
[11] Cheng SY. et al.: Development of Cosme-tic Textiles Using Microencapsulation Tech-nology. RJTA 12 (2008) 4, 41-51
[12] Hong K., S. Park: Melamine resin micro-capsules containing fragrant oil: synthesis and characterization. Mater-ials Chemistry and Physics 58 (1999) 128-131
[13] Hong K., S. Park: Preparation of poly(L-lactide) microcapsules for fragrant fiber and their characteristics. Polymer 41 (2000) 4567-4572
[14] Passarini N. et al.: Solid Lipid Microparticles Produced by Spray Congealing: Influence of the Atomizer on Micro-particle Characteristics and Mathematical Modelling of the Drug Release. Journal of Pharmaceutical Sciences 99 (2010) 2, 916-931
[15] Jyothi NVN. et al.: Microencapsulation techniques, factors influencing encapsulation efficiency, Journal of Microencapsulation 27 (2010) 3, 187-197
[16] Lee-Fong S., O. Chee-Sian: Effect of pH on Garlic Oil Encapsulation by Complex Coacer-vation, J Food Process Technol 4 (2013) 1, 1-5
[17] Gouin S.: Microencapsulation: industrial appraisal of existing technologies and trends, Trends in Food Science & Technology (2004) 15, 330-347
[18] Alvim ID., CRF. Grosso: Microparticles obtained by complex coacervation: influence of the type of reticulation and the drying process on the release of the core material, Ciência e Tecnologia de Alimentos 30 (2010) 4, 1069-1076
[19] Xiao J.-Xia et al.: Microencapsulation of sweet orange oil by complex coacervation with soybean protein isolate/gum Arabic, Food Chemistry 125 (2011) 1267-1272
[20] Perignon C., G. Ongmayeb: Interfacial polymerization versus cross-linking microencapsulation, Bioencapsulation Innovations, March (2013) 8-10
[21] Oliveira WP. et al.: Food and herbal products. In: Spray Drying Technology 1. Singapore (2010) pp. 132-133
[22] Training Papers Spray Drying. www.uechi-prd-web.unic24.net (December 2013).
[23] Li YO.: Development of microencapsulation-based technologies for micronutrient fortification in staple foods for developing countries, PhD Thesis, University of Toronto, Canada, 2009
[24] Nelson G.: Microencapsulation in textile finishing. Rev. Prog. Color. 31 (2001) 57-64
[25] Li S. et al.: UV curing for encapsulated aroma finish on cotton, Journal of The Textile Institute 96 (2005) 6, 407-411
[26] www.euracli.fr
[27] www.avonpolska.pl
[28] www.nordcell.pl