Pressure-sensitive impermeable microcapsules for controlled release of essential oils

E. Knez[1] , B. Boh[2]

[1]Aero Chemical, Graphic and Paper Manufacturers, Celje, Slovenia (emil.knez@aero.si), [2] University of Ljubljana, Slovenia (bojana.boh@ntf.uni-lj.si)

Introduction

Industrial uses of essential oils are limited by their high volatility. Microencapsulation technology offers a solution to achieve reduced volatility of essential oils, controlled release under desired conditions, as well as applications on different carriers, such as scented antimicrobial textiles, and "scratch and sniff" papers. In our research, aminoaldehyde microcapsules were produced with impermeable pressure-sensitive walls to achieve protection and controlled release of essential oils by mechanical pressure.

Materials and methods

Natural essential oils of *Citrus sp.*, *Eucalyptus sp.*, *Lavandula sp.*, *Melaleuca alternifolia*, *Rosmarinus officinalis*, *Salvia officinalis*, and synthetic aromatic mixtures of rose and strawberry were used as antimicrobial and/or perfume agents. Oils were diluted with isopropylmyristate to conc. 60-70% for scented papers, and to conc. 25% for textile applications. Microcapsules were prepared by *in situ* polymerisation of aminoaldehyde resins (Knez, 1995; Kukovič & Knez, 1997) in a 10L and 200L reactor. Non-woven 250g/m^2 polyester textile was impregnated by a liquid formulation, consisting of aqueous microcapsule suspension and acrylic latex binder. The impregnation machine contained a textile carrier transport through the impregnation basin, and a drying/curing channel at T = 120° - 140 °C. The end product contained 150g of dry microcapsules per m^2 of textile. For the production of repositionable scented paper notes, an aqueous formulation containing microencapsulated perfumes with an acrylic viscosity modifying agent was printed on a 80g/m^{2}paper carrier (1.0-1.5 g dry weight of microcapsules per m^2 of paper) on an industrial flexographic printing machine, together with the microsphere containing adhesive. Paper was cut and assembled into repositionable paper notes.

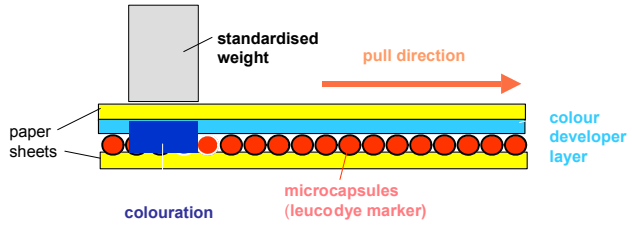

***Fig. 1**: Smudging colouration test for the evaluation of microcapsule wall resistance to mechanical stress,*

Mechanical testing of pressure-sensitive microcapsules was performed in a combination of microencapsulated leuco dye and colour developer (standard smudging colouration test, Fig. 1). Textile shoe insoles were tested by walking (experimental person 80 kg, average 3 km per day, total distance of 150 km). Thermal properties of microcapsules were tested by an accelerated test of diffusion at 120°C (modified smudging colouration test at elevated temperature). Headspace gas chromatography was used for a quantitative determination of essential oil components in microcapsules and in final products, using internal standards (flame-ionisation detector, capillary column HP Ultra 1 and HP Carbowax M20, initial temperature 60°C, temperature gradient 2.5°C/min, final temperature 190°C). The microcapsule size and size distribution were measured by Alkatel Cilias Laser Granulometer 715.

Results and discussion

Impermeable pressure-sensitive microcapsules containing mixtures of essential oils were successfully produced at an industrial scale (Fig. 2,3).

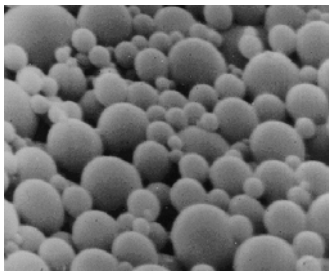

Fig. 2: Microencapsulated essential oils on a paper carrier

Fig. 3: Textile impregnated with essential oil microcapsules

Microcapsules were mechanically and thermally resistant to protect the essential oils during the production process of impregnating and drying of textiles, and of flexographic printing on papers.
In the accelerated diffusion test, components of essential oils diffused through the microcapsule wall at different rates, depending primarily on the concentration and partial vapour pressure (Tab 1).

	concetration			rel loss (%)	
Essential oil component	start	after 2	4 hour	after 2	4 hour
α-pinene	1,04	0,94	0,30	32,1	83,0
camphene	0,75	0,77	0,58	22,9	53,7
β-pinene	0,50	0,44	0,14	32,8	83,6
mircene	0,18	0,07	0,00	72,2	100,0
limonene	0,58	0,46	0,11	40,5	89,0
1,8 cineole	3,53	3,61	3,90	23,3	34,1
thujone	1,84	1,76	1,69	28,4	45,3
camphere	3,19	3,11	3,89	26,8	27,3
linalool	5,49	4,05	0,98	44,7	89,4
linalyl acetate	0,94	0,78	0,28	37,7	81,9
terinen-4-ol	1,41	0,81	0,13	57,0	94,4
borneol	1,62	1,48	1,15	31,7	57,8
i-propylmyristate	75,16	76,99	80,66	23,2	36,0
Other components	3,78	4,74	6,21	6,0	2,0
Total	100	100		**25,3**	**40,3**

***Table 1:** Accelareted test of diffusion at 150 °C - microcapsule composition analisys by gas chromatography*

Particle size and distribution (Fig. 4), wall thickness and pore structure of walls had a great influence on mechanical and thermal properties of microcapsules. These were altered by process parameters: ratio of emulsifier/core, revolution of a mixer diameter, pH value, temperature and time of polymerisation, and concentration of the oil phase.

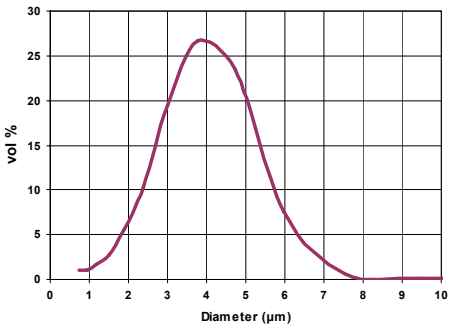

***Fig. 4:** Microcapsule size distribution (mean diameter 3.60 µm; wall thickness 0.07 µm. Dry microcapsules contained: core 75%, wall 14%, emulsifier 10%)*

Mechanical tests by walking proved that shoe insoles contained some unbroken microcapsules even after 150 km of walking. The result was confirmed also by a headspace chromatographic test. After 50 km of walking, shoe insoles contained 62 - 72% of microencapsulated active ingredients. The release was more intense on insole parts exposed to higher mechanical pressure. Antimicrobial activity test confirmed that a selected mixture of essential oils possessed a strong antibacterial and fungicide activity, wich was substantially prolonged by microencapsulation..

Conclusions

Microencapsulation provided an effective protection of essential oils during the industrial production of antimicrobial textiles and scented papers, as well as during the storage of final products – antimicrobial shoe insoles and repositionable scented paper notes. Under defined conditions, essential oils were readily released by a mechanical pressure (walking, scratching, and writing). Microencapsulation process by in situ polymerisation of aminoaldehyde resins was adapted to a diversity of different batches of essential oils, as well as to achieve the desired level of permeability and mechanical resistance of microcapsule walls.

Acknowledgement

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References

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