Microencapsulation of peppermint oil during spray-drying

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Introduction

Essential oils are natural liquid products obtained from plants by hydro- or steam distillation. Oils contain from dozens to hundreds of components – secondary plant metabolites. The essential oils are used as natural aromas in food and toiletry, and due to medicinal properties, many of them are used in conventional medicine and aromatherapy. Components of the essential oils are first of all mono- and sesquiterpene hydrocarbons and their oxygenated derivatives, and also phenylpropanoids. These are volatile substances sensitive to oxygen, light, moisture and heat. Stability of essential oils can be much increased by using microencapsulation. This process not only protects against losses and chemical changes during food production and storage, but also enables production of aromas in the form of powders – products with new properties. Main advantages resulting from their application include a much facilitated incorporation into solid products and a possibility of controllable release of the aroma during consumption.
Dry microencapsulation product in the form of powder is most often obtained in spray drying of a multicomponent suspension, although other methods, e.g. extrusion, freeze drying, or co-crystallization are used as well [1]. As carriers for aroma microencapsulation carbohydrates are mainly used, i.e. starch and its derivatives, e.g. maltodextrin [2,3], β-cyclodextrin [4,5] and gum [6,7]. Recently, also proteins, e.g. milk or soya proteins, have become popular [6,8]. Frequently, mixtures of the mentioned materials are used in different proportions [8,9].

Materials and Methods

A material for the microencapsulation of essential oils was maltodextrin DE 16 of class "S" obtained from PPZ "NOWAMYL" S.A. at Łobez. Peppermint oil (*Mentha piperita*) is produced by steam distillation of peppermint leaves, and its main components are oxygenated monoterpenes: alcohols, esters and ketones.
Emulsions were formed in a flow system with the use of a UP200S ultrasonic homogenizer (IKA – Dr. Hielscher, GmbH, Berlin). Tween®80 surfactant (Fluka) and a relevant amount of essential oil was added to an aqueous maltodextrin solution while mixing steadily. The solution was then emulsified in a closed system of the flow homogenizer with a cooling jacket. In about 5 h of the emulsification process, the suspension was flowing ca. 35-40 times through the chamber equipped with an ultrasonic probe. All tests were made for solutions with maltodextrin to water ratio 30:70 and essential oil content 10, 20 and 30 wt.% in relation to maltodextrin, with the addition of Tween®80 equal to about 0.1 wt.% in relation to maltodextrin.
The emulsion was dried in a SR 16 spray dryer (METALCHEM), equipped with a high-speed disk atomizer of rotation velocity ca. 22000 min^{-1}. The drying chamber diameter was 1.5 m, total dryer height about 2.5 m. Drying was performed in the concurrent phase flow, at inlet air temperature ca. 150°C and outlet air ca. 80°C.
During analyses the following properties of produced powders were determined: moisture content, absolute powder density, (equilibrium) sorptivity of water vapor in the powder, microparticle size, total content of essential oil in the product and on microparticle surface (in this way encapsulation degree was estimated), chemical composition of essential oils used and retained in the product.

Microscopic photographs of the material were also taken. Essential oil content in emulsion and microencapsulated product was estimated by hydrodistillation using Deryng apparatus for 3 h (according to Farmakopea Polska VI, 2002). Analysis of essential oil composition on the product surface was made by the HS-SPME method (headspace – solid phase microextraction). 2.0 g of powder was placed in a 10 ml glass vial and allowed to equilibrate for 15 min at 60°C. After this time, the Supelco SPME fiber coated with Car/DVB/PDMS was exposed to the headspace for 30 min at 60°C and than the fiber was transferred to the injection port of the GC system.
The components of essential oils were separated by gas chromatography (GC) method using a MEGA 5300 apparatus (Carlo Erba Instruments, Rodano, Italy), with a flame ionization detector (FID) and SSL injector, with the application of a CP Sil 5 CB capillary column (Chrompack) 30 m ×0.32 mm, film thickness 0.25 μm. Temperature of the column was programmed from 50 to 300°C, rise rate 4°C/min, injector temperature 320°C, detector temperature 310°C, nitrogen flow rate 1.0 ml/min. The content of components was determined by the inner normalization method.
Coupling of gas chromatography with mass spectrometry (GC-MS) analysis was made using a MD800 mass spectrometer coupled with Fisons GC 8000 gas chromatograph. Conditions of the chromatographic analysis: column type and analytical conditions as in GC, carrier gas – helium, flow rate 0.8 ml/min. Parameters of the mass spectrometer: ionization energy 70 eV, ion source temperature 200°C. The essential oil components were identified by comparing: retention indices (RI) relative to a series of alkanes (C_7-C_{26}) with standards, and MS spectra with standards or NIST, LBTX and MassFinder 3 databases and with literature [10].
For the determination of absolute material density AccuPyc 1330 Helium Pycnometer (Micromeritics, USA) was used. Equilibrium moisture content was determined as a sorption of water vapor in a NOVASINA (Axair Ltd., Pfaffikon, Switzerland) instrument type A_w SPRINT.

Results and Discussion

Experiments were made at constant conditions of emulsion preparation and spray drying - Table 1. Table 1 gives also results of analysis of many properties of drying and microencapsulation products.

Emulsion	Temperature inlet / outlet [°C]	Density [g/cm^3]	Moisture content X or b [kg/kg d.b./ wt.%]	Oil content in the product [%]	Process yield [%]	Oil content in the powder after 5 months [%]
Maltodextrin	160 / 80	1.4209	0.043 / 4.10	-	-	-
M-10	150 / 80	1.4260	0.030 / 2.88	7.1	70.6	6.4
M-20	149 / 82	1.3354	0.0173 / 1.70	13.5	67.5	12.8
M-30	150 / 80	1.2904	0.0191 / 1.875	18.2	57.2	18.0

Table 1. Drying conditions and product characteristics

It follows from the analysis that the applied drying parameters, and especially a relatively low air temperature at the dryer inlet, are sufficient to produce powder of low final moisture content – in most cases below 3 wt.%. It can also be considered that the composition of emulsion being dried had no important effect on moisture content in the product. The determined absolute density of powders shows that it depends on the content of oil in powder – with an increase of oil hold-up (encapsulation) in maltodextrin, the powder density decreases. This rule is not confirmed only in the case of M-10 product, i.e. the product of drying of the 10 wt.% peppermint oil emulsion. The determined value is higher than the density of pure maltodextrin, which can be within the determination error.
Efficiency of the microencapsulation process was estimated by determination of the total essential oil content in the product and comparison with the initial oil to maltodextrin ratio. Results are given in Table 1. The content of peppermint oil in the powder produced from the emulsion that contained

10, 20 and 30 wt.% of this oil in relation to maltodextrin, was 7.1, 13.5 and 18.2%, respectively. Efficiency of the process of microenencapsulation decreased with an increase of the initial oil concentration from 70.6% for 10 wt.% oil to 57.2% for 30 wt.% oil.
The durability of microencapsulated complex - peppermint oil in maltodextrin was examined after 5 month storage in not tight glass containers under ambient conditions. The results are presented in the last column of Table 1. Apparently only several percent of the initial quantity of peppermint oil in powder was lost.
Next, the chemical composition of initial peppermint oil was compared with the composition of this oil as a suspension in the prepared emulsion, encapsulated in the product and held-up on the powder surface for variants with 30 wt.% oil content– results are given in Table 2. The quality and quantity of the oil suspended in the emulsion and encapsulated in the powder was the same and differed only slightly from the initial oil. This confirms that the most volatile components, i.e. monoterpene hydrocarbons and menthone, had been partially evaporated while preparing the emulsion. The quantitative composition of the surface oil was much different: the percentage of monoterpene hydrocarbons and menthone decreased more than twice, on the other hand the content of menthol increased by ca. 30% and menthyl acetate by about 50%. Main components of the peppermint oil are oxygenated monoterpenes, so their better encapsulation ability is obvious.

Component	Initial oil %	Oil in emulsion %	Oil in powder %	Oil on powder surface, %
limonene	1.96	1.71	1.68	0.76
menthone	**22.96**	**21.00**	**21.19**	**9.21**
isomenthone	8.84	8.30	8.60	6.24
neomenthol	6.30	6.47	6.67	6.14
menthol	**50.07**	**54.00**	**53.30**	**70.21**
isomenthol	0.24	0.15	0.26	0.22
menthyl acetate	3.53	3.73	3.68	5.41
isomenthyl acetate	0.10	0.10	0.09	0.10
monoterpene hydrocarbons	**2.74**	**2.45**	**2.36**	**ca. 1**
oxygenated monoterpenes	**95.04**	**95.69**	**95.74**	**97.67**
sesquiterpene hydrocarbons	**2.12**	**1.86**	**1.86**	**ca. 1.3**

Table 2. Composition of peppermint oil (30 wt.% of the oil)

Figure 1 shows isotherms of water vapor sorption by different products of drying and micro-encapsulation determined at the constant temperature of 25°C, and Figure 2 shows isotherms of water vapor sorption by the M-20 product of drying and microencapsulation determined at different temperatures. From these curves we can deduce a very clear effect of the content of oil in the powder on sorptivity and also on hygroscopicity of the dried product. Equilibrium curves of powders with essential oil are much below the curve for pure maltodextrin dried in the same conditions. Hence, it can be stated that the product of microencapsulation is less sensitive to water vapor in the environment (i.e. more stable) at medium air humidity than the pure carrier. However, all products were dissolved completely, or rather “vitrified”, at the same ambient humidity, i.e. relative humidity of ca. 0.75. At the higher temperature sorptivity of the same encapsulated powder product M-20 is lower – Figure 2.

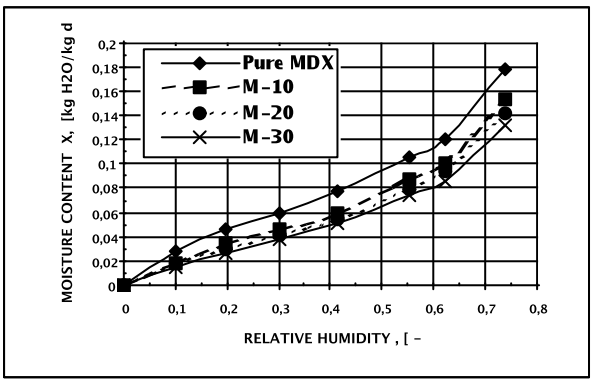

Figure 1. Equilibrium moisture content of powders obtained by spray drying. Sorption temperature 25°C.

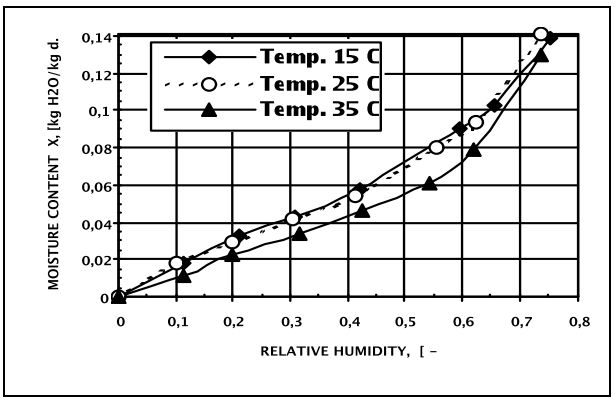

Figure 2. Equilibrium moisture content of M - 20 powders obtained by spray drying. Different sorption temperatures.

Conclusions

Applicability of spray drying of multicomponent emulsions of essential oils in the process of their microencapsulation in a carrier material was confirmed. Microencapsulation in maltodextrin is a suitable process to obtain powdered peppermint oil characterized by high durability. Process efficiency can be probably increased due to modification of the conditions in which emulsions are prepared and dried.

The encapsulation of substances with low boiling points requires a more precise determination of process conditions, especially at the stage of preparing the emulsion.

Results of preliminary investigations are promising and offer a basis to extend the scope of research, among the others, on other carrier materials and essential oils.

It is worth noting that the equipment can be used in the investigation of a variety of multicomponent systems that can be spray-dried in a broad range of process variables.

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