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Industry-scale spray-drying microencapsulation of orange aroma

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A b s t r a c t. The article describes an attempt to solve a real industrial problem, connected with the not very efficient enclosure of orange aroma during industrial microencapsulation, by the replacement of conventional mixing by single-stage homogenization during feed emulsion preparation. The powders obtained from solutions after 17 MPa homogenization were characterized by a lower water content and better flowability. The powders from solutions after 25 MPa homogenization showed better myrcene retention. An additional aim of this work was to investigate the physicochemical properties of powders obtained from the cyclone container and the drying chamber. This approach is connected with the fact that older generation spray dryers do not have systems protecting against local powder deposits in the chamber. In such situations, in order to reduce losses, the powder from the cyclone container and the one from chamber are mixed together, even if they have different properties. The results obtained show the significant differences between the powders collected from the cyclone container and the chamber of the spray dryer in terms of water content, bulk density, particle-size distribution and aroma retention. The powders obtained from the chamber were characterized by a lower water content, better flowability and a lower porosity value. Hierarchical cluster analysis confirmed the differences between the investigated microcapsule variants.

K e y w o r d s: powders, aromas, spray-drying, microencapsulation, aroma retention

INTRODUCTION

The importance of the role of smell in the perception of the outside world by people was confirmed by the winners of the Nobel Prize in 2004, Linda B. Buck and Richard Axel, who found that olfactory receptors are based on over 1000 different proteins that are encoded by a total of 3% of our genes. Such a mechanism makes it possible to recognize over 10 000 smells, whereas by using language, we can recognize only five tastes – salty, sweet, sour, bitter and umami. Physiology determines the role of fragrances in the food industry (Breer, 2003; Buck, 2004). The nature of the aroma added to the product often determines its acceptance or rejection by the consumer. The aromatic compounds are easy to evaporate, oxidize and decompose, so it is important to choose the best way to "pack" and preserve them, for example by microencapsulation using the spray-drying method. The spray-drying process is one of the most popular microencapsulation methods in the food industry. It is economical, uses easily available equipment and produces good quality powders (Reineccius, 2004; Jafari *et al*., 2008; Feenades *et al*., 2014).

Orange flavours are some of the most popular flavours. They are widely used in both the food and pharmaceutical industries. They are added to sweets, cakes, jellies, gels, teas, drinks, medicines and diet supplements. The most important component of the orange flavour is "sweet orange oil", which is often almost 95% of the whole aromatic portion of the flavour. Its annual production exceeds 50 thousand tons, which represents the bulk of all essential oils manufactured. It is mainly extracted from orange peel, which is a waste product of juice production, so it is relatively cheap. Apart from their characteristic antimicrobial properties, citrus oils also have a calming effect. However, their basic application is in the food flavour industry. They are commonly added to soft drinks, sweets, cakes and desserts. On the other hand, bitter orange oil is widely used in refreshing drinks, liqueurs and confectionery. Depending on the other ingredients of

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the aromatic portion, orange flavours may have notes of butter, candy, citrus, floral, fruit, green, tea or vanilla. For example, a small addition of geraniol provides floral or rose notes, hexane – green, myrcene – tea, octanal or decanal – citrus-skinned, and maltol – candy (Brud and Konopacka-Brud, 2009; Dorland and Rogers, 1977; Wright, 2004).

The production of powdered citrus aromas is particularly difficult due to the high content of citrus oils, which precipitate from the carrier solution. According to McClements (2005) emulsion stability is a very important factor in the microencapsulation of flavours and oils, because they are generally insoluble in water. However, this results in an ineffective enclosure of the aroma in the microcapsule and in the poor stability of the finished product. The taste of improperly microencapsulated citrus aromas during the storage becomes bitter. In addition, the delaminating emulsion causes differences in colour and taste between production batches. The emulsion-feed solution for the spray-drying process should be stable throughout the whole microencapsulation process, including short-term storage before drying, transportation to the spray disc and the entire drying time (Thies, 2004).

The type and the parameters of the homogenization process change the properties of the emulsion , which further affects the physicochemical properties of powders after microencapsulation (Frascareli *et al*., 2012). The objective of emulsification is to produce uniform and small droplets during subsequent atomization in a spray-drying chamber (Jafari *et al*., 2008; Garcia *et al*., 2012). The droplet size may be reduced by increasing the pressure and the time of homogenization (McClements *et al*., 2007; Janiszewska *et al*., 2015). However, on the other hand, there are also examples that indicate that overly intense homogenization may increase the size of the droplets (Floury *et al*., 2003; Jafari *et al*., 2007).

The main aim of the research was to check whether the replacement of mixing by single-stage homogenization before spray-drying microencapsulation would improve the quality of the powders in terms of 1) better retention of aromatic ingredients, resulting in an increased intensity of the aroma and possible dosage reduction of the finished product, 2) improved physical properties, *i.e.* flowability.

An additional aim of this work was to investigate the physicochemical properties of powders taken from different locations of the spray dryer (cyclone container and chamber). Many Polish companies have older spray dryers that are not equipped with systems to prevent local powder deposits in the chamber (*e.g.* hammers, air brushes). In such dryers, in order to reduce losses, the powders from the receiver and the chamber are mixed together.

MATERIALS AND METHODS

The orange aroma was produced by the "Pollena-Aroma" company (Warsaw, Poland), where the industrial microencapsulation process was also carried out. The real complex aromas, manufactured by the "Pollena-Aroma" company were analysed for the article. The recipes were reserved by the manufacturer. Maltodextrin (DE 15) derived from Pepees S.A. (Łomża, Poland), and gum arabic from Nexira Food (Rouen, France) were used as wall materials (carriers) for the microencapsulation process.

The solid concentration of feed aroma/carrier solutions was 50% w/w, the ratio of the aromatic part to the carrier was 1:4 (the carrier was a mixture of MD and GA at the ratio of 1:7). 3 different types of feed solution pretreatment were applied: mixing (1500 rpm) by mechanical stirrer (M), mixing by mechanical stirrer (1500 rpm) and then homogenization at 17 MPa (H17), mixing by mechanical stirrer (1500 rpm) and then homogenization at 25 MPa (H25).

The microencapsulation process was carried out under industrial conditions at the "Pollena-Aroma" company. A NIRO spray dryer (A/S, Germany), equipped with a co-current, double-canal atomization nozzle was used. The dimensions of the dryer were 2×2.7 m (diameter×height). Process parameters: air inlet temperature 180°C, air outlet temperature 80°C, air flow rate 100 g s^{-1}, feed rate 8.3 g s^{-1}. The powders were obtained from two production batches, and were collected from two locations in the spray dryer – the cyclone container (C – powder collected at the bottom of the cyclone), and dryer chamber (Ch – powder which did not pass from the chamber to the cyclone container – to obtain this powder the dryer walls were tapped with a hammer and the powder was collected by opening the bottom of the chamber).

The aroma compounds of the feed solutions and the powders obtained were analysed using Headspace GC-MS (GC Agilent 7890A, Headspace Sampler 7697A) with Identification System (NIST). The powders were diluted in water to achieve the same amount of dry matter in the tested sample, as in the samples of the feed solutions. Samples were heated to 80°C and then 1 mL was injected automatically to the column (Agilent column HP-1). The analysis conditions were: oven temperature program: (1) 50°C 5 min^{-1}, (2) 50-200°C – 2°C min^{-1}, (3) 200-250°C – 5°C min^{-1}, (4) 250°C 10 min^{-1}; FID temperature – 300°C; injector temperature – 250°C; helium flow rate 1.9 mL min^{-1}. The content of aromatic compounds in the samples of reconstituted powders was compared to the content of these compounds in the samples of feed solutions containing the same amount of solids (the samples of feed solutions were treated as initial samples, in which the aroma compounds content was assumed to be 100%). The percentage of the content of aromatic compounds in powders was calculated based on peak areas comparison (reconstituted powders vs. feed solution). Thus, the percentage of aroma compound in the reconstituted powder was related to the initial content of each component of the feed solution (taking into account that the solids content in the tested samples was the same).

The water content (wet-basis, WC) was measured by drying at 105°C for 4 h. The bulk density was measured by using an automatic volumeter STAV 2003 (Engelsmann

AG, Germany): loose bulk density ρ_L and tapped bulk density ρ_T, by tapping 100 times. The apparent density ρ_P of the particles was measured using a helium pycnometer Stereopycnometer (Quantachrome, Boynton Beach, USA). Based on bulk densities and apparent density the following calculations were performed: porosity of loose bed $\in_L = 1-(\rho_L/\rho_P)$, porosity of tapped bed $\in_T = 1-(\rho_T/\rho_P)$, and Hausner ratio $HR = \rho_T/\rho_L$. The microphotographs of the powder particles were taken using a TM-3000 Tabletop Scanning Electron Microscope Hitachi (Tokyo, Japan). At least 200 particles of every sample powder were outlined in the microphotographs using the MultiScan program (Computer Scanning Systems, Warsaw, Poland), and the sieve diameters of the particles were provided automatically by the software.

The glass transitions of the powders were measured using differential scanning calorimetry with a DSC Q200 (TA Instruments, New Castle, USA). The calibration was carried out on indium, an empty pan was used as a reference. 4-5 mg samples were weighed into pressure pans which were then heated according to the following program: (1) 20°C 2 min^{-1}, (2) 20-150°C – 10°C min^{-1}, (3) 150°C 1 min^{-1}, (4) 150-20°C – 10°C min^{-1}, (5) 20°C 1 min^{-1}, (6) 20-150°C – 10°C min^{-1}. The glass transition of the first heating cycle T_{g1} and of the second cycle T_{g2}, were evaluated using Universal Analysis software (TA Instruments, New Castle, USA).

Powders reconstituted in water were evaluated using the duo-trio method. The analysis was carried out by a six-person qualified staff of the "Pollena-Aroma" company.

The results were statistically analysed using STATISTICA v.13 software (Dell Inc., USA). A one-way analysis of variance and Tukey HSD test were used (at significance level $\alpha = 0.05$). In the case of a failure to fulfil the assumptions of variation connected with the heterogeneity between the compared groups, a t-Student test (in the case of the exclusion of one group) or Kruskala-Wallis test (in the case of the exclusion of more than one group) was used. In order to analyse the interrelationships between the determined parameters of the multivariate statistical methods: a principal component analysis (PCA), and a hierarchical cluster analysis (HCA) were also performed. The PCA was used to obtain a small set of principal components (PC) that explain most of the variability within the data set. HCA was based on Euclidean distance to calculate the sample similarities and to indicate the complete linkage clustering by Ward's algorithm.

RESULTS AND DISCUSSION

α-pinene, myrcene, limonene, and linalyl acetate, commonly used in orange flavours, are responsible for their characteristic note. Limonene, the main ingredient of orange oils, is characterized by an intense citrus aroma. α-pinene in pure form suggests the smell of pine and fir trees. Myrcene in a pure form has a smell of cloves, while linalyl acetate is characterized by a delicate floral note. It constitutes 65% of petitgrain oil, which is obtained from leaves, twigs and small unripe fruit of bitter orange, by a distillation method (Dorland & Rogers, 1977; Wright, 2004).

The retention of the individual aromatic compounds in the obtained microcapsules after spray drying is presented in Table 1. It ranged from 56.3±1.3 to 93.1±3.9%, and depended on the type of the compound, the method applied for emulsion preparation, and the location of powder collection. Similar levels of retention were reported previously i.a. by Yuliani *et al.* (2006): after the microencapsulation of D-limonene with starch and β-cyclodextrin by extrusion, the retention of D-limonene was 92.2%. In the work presented by Chen *et al.* (2013) the retention of limonene after spray-drying microencapsulation was 80.5±2.5%, while after the use of the freeze-drying method it was 54.0±1.7%. Fisk *et al.* (2013) using spray dried sunflower seed oil, with modified starch as a carrier, noted the retention of D-limonene in the range 55-59%. Most scientific articles regarding flavour microencapsulation concerns the microencapsulation of individual compounds, essential oils or model group of several aromatic compounds, on a laboratory scale. The powders analysed for the article, produced by the "Pollena-Aroma" company, contained a dozen or even several dozen different compounds. The industry flavour compositions consist of a large number of ingredients, because they should give the product a specific, sophisticated fragrance, often difficult to imitate and standing out from the competition (clearly associated with a specific product).

Table 1. Percentage of individual aromatic compounds in the powders compared to the content in the feed solutions (initial 100%): after mixing from the cyclone container (MC) and from the chamber (MCh); after mixing and homogenization at 17 MPa from the cyclone container (H17C) and from the chamber (H17Ch); after mixing and homogenization at 25 MPa from the cyclone container (H25C) and from the chamber (H25Ch)

Variants	α-Pinene	Myrcene	Limonene	Linalyl acetate
MC	61.1±6.4^{ab}	74.7±6.6^{ab}	89.8±8.4^{a}	81.7±32.4^{a}
MCh	60.2±3.7^{a}	61.6±4.2^{a}	90.0±6.5^{a}	74.8±25.3^{a}
H17C	68.5±7.1^{bc}	78.8±5.6^{bc}	93.1±3.9^{a}	73.7±26.6^{a}
H17Ch	56.3±1.3^{a}	72.7±6.5^{ab}	88.5±4.8^{a}	76.9±19.0^{a}
H25C	70.9±5.0^{c}	78.6±6.5^{c}	87.9±12.6^{a}	75.1±30.5^{a}
H25Ch	61.1±4.2^{ab}	75.9±9.9^{bc}	90.1±7.6^{a}	80.1±32.2^{a}

a-d – differences between mean values followed by different letter in columns were statistically significant ($p < 0.05$).

The highest retention was presented for limonene, then linalyl acetate and myrcene, while the smallest retention was observed for α-pinene. In the case of α-pinene and myrcene the higher retention was found in powders taken from the cyclone container, compared to the powders from the chamber. In all probability, the high temperature in the chamber caused an escape of volatile compounds. The powders obtained from H17 and H25 emulsions had a higher retention of myrcene, compared to powders obtained from

M emulsions. Higher pressure homogenization probably reduced the delamination of the emulsion and also reduced the amount of the oils outside of the microcapsules. The same result – an increase in oil retention with increasing homogenization pressure before microencapsulation through the spray-drying method, was observed by Garia *et al*. (2012) and Soottitantawat *et al*. (2003).

The retention of limonene and linalyl acetate was higher than the results observed for α-pinene and myrcene, which could be caused by lower volatility, which, in turn, results from the higher molar mass of the former two compounds (Nivaldo, 2017). There was no effect of the emulsion preparation method and the powder collection location on the retention of limonene and linalyl acetate.

The powders obtained had a spherical shape with an irregular, dented surface. Fernandes *et al*. (2008) and Soottitantawat *et al*. (2003, 2005) obtained a similar morphology for particles of D-limonene (Fig. 1). As may be seen in the microphotographs, powders taken from the chamber were characterized by larger particles than the powders from the cyclone container.

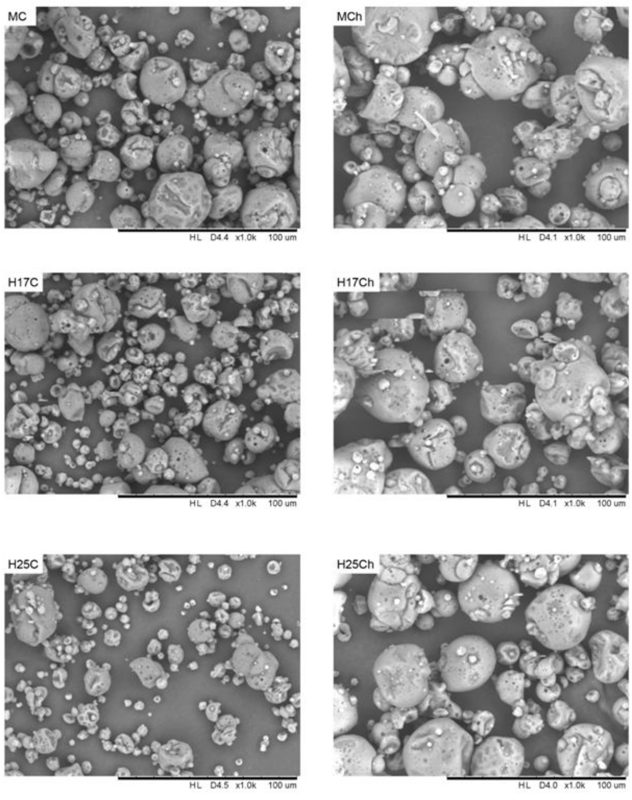

Fig. 1. Microphotographs of orange powders (mag. 1000×).

Analysing the most frequent diameters, it was found that powders obtained from the mixed solutions were characterized by the largest particle size: 5 (MC) and 7 µm (MCh), while powders obtained from solutions after 17 MPa homogenization – 5 (H17C) and 10 µm (H17Ch) respectively but powders from solutions after homogenization of 25 MPa – 3 (H25C) and 9 µm (H25Ch) respectively (Fig. 2). By analysing the most frequently occurring diameters, it may be concluded that the powders from the cyclone container were characterized by a smaller particle size than the powders from the chamber. Ordonez and Herrera (2014), who microencapsulated limonene by spray drying, obtained particle sizes in a range from 1 to 9 µm with the use of arabic gum as a carrier, and 4-17 µm when a combination of arabic gum and cassava starch in the proportion of 1:1 was applied.

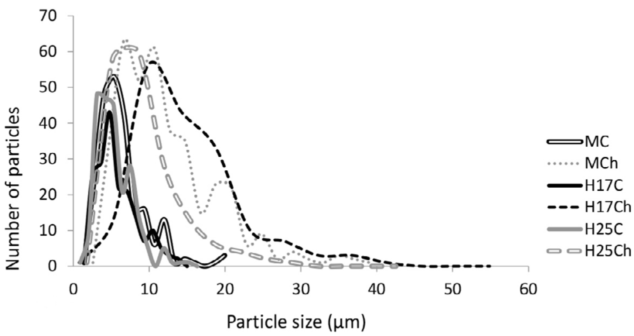

Fig. 2. Particle-size distribution of microencapsulated orange aromas.

The particle-size distribution curves (Fig. 2) were characterized by multiple peaks, which may be considered as a beneficial phenomenon – when smaller particles have the possibility to fill the spaces between the larger ones the powder bed takes up less volume (Silva *et al*., 2013). It was observed, that powders from the chamber were characterized by a wider particle-size distribution.

From the cumulative curves (Fig. 3), it may be concluded that particles with a diameter of less than or equal to 10 µm reached 89 (MC) and 51% (MCh) of the powder particles from the mixed solutions taken from the cyclone container and the chamber respectively, 91 (H17C) and 42% (H17Ch) of the powder particles from solutions after homogenization 17 MPa and 96 (H25C) and 75% (H25Ch) powder particles from solutions after 25 MPa homogenization. Thus, the powders from the cyclone container were characterized by a smaller particle size than the powders from the chamber. According to Reineccius (2004) for the dryers that give operators the option of collecting powder from the bottom and side, larger particles are collected from

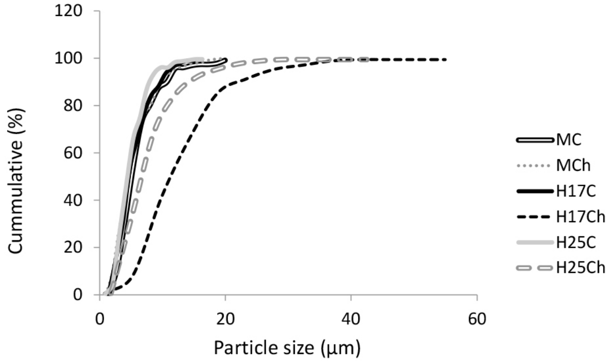

Fig. 3. Cumulated curves of particle-size distribution of microencapsulated orange aromas.

the bottom of the dryer, while finer powders are collected from the side (after the cyclone). This effect may be explained in two ways: on the one hand, it is most likely that the smaller particles were blown out with outlet air to the cyclone more easily than particles of a higher magnitude. On the other hand, powders from the chamber could be larger due to the intensive evaporation of water in the chamber causing a greater internal porosity (confirmed by the lower particle density of the chamber collected powders) of the powders. Furthermore, larger particles have a greater inertia, they collide with the chamber wall and form a deposit.

No effect of the solution pre-treatment on the particle size was found. During the microencapsulation of basil essential oil Garcia *et al*. (2012) observed that powders obtained from the emulsions homogenized at different pressures, with constant oil concentrations, also had similar particle sizes.

It is necessary to achieve a low water content in microencapsulated powdered aromas in order to attain sufficient microbial stability and long shelf life – moist particles have a tendency to agglomerate and this causes "the escape" of active compounds, caking and reduces flowability (Silva *et al*., 2013). The obtained values of the water content (from 2.6±0.0 to 4.5±0.0) were in a range typical for spray-dried powders (Table 2). Islam (2016) obtained lower values of water content (2.29-3.35%), for orange juice powders produced by pressurized spray dryer. This was explained by the better conditions of heat transfer of the superheated steam (at a temperature of 200°C), compared to hot air (used in traditional spray drying).

The powders collected from the drying chamber had a lower water content, compared to those from the cyclone container, but the differences were not statistically significant. In previous studies carried out both on a laboratory and on an industrial scale (Jedlińska *et al*., 2013; Jedlińska *et al*., 2014) it was found that powders from the cyclone container had a statistically higher water content than the powders from the chamber. This result was explained by long-term exposure at high temperatures in the chamber.

Among the powders obtained from the cyclone, the highest water content was measured in a premixed MC sample, as well as from among powders from the chamber (MCh). Jafari *et al*. (2007) also found that a decrease in the water content of spray dried d-limonene microcapsules occurred after the change in the mixing parameters for high pressure homogenization. Janiszewska *et al*. (2011), who spray dried lemon aroma with the addition of arabic gum, did not notice any significant differences between the water content of powders obtained from high-speed mixing solutions (24000 rpm) and single pressure homogenization (30 MPa), while statistically significant increases in the water content were found by using two-stage pressure homogenization (30/10 MPa). When spray drying essential oil, Garcia *et al*. (2012) did not observe the impact of emulsion viscosity, homogenization pressure and oil concentration on moisture content.

The loose bulk density of the powders collected from the cyclone container and the chamber of the spray dryer was 0.39±0.02 (C) and from 0.40±0.01 to 0.43±0.02 g cm^{-3} (Ch), respectively (Table 2). For comparison, the bulk densities obtained by Flores-Martines *et al*. (2004), who spray dried orange oil with different levels of gum arabic addition (15, 20, 25%), were lower (0.19-0.22 g cm^{-3}). In contrast, the bulk density of the orange juice powder was between 0.41 and 0.85 g cm^{-3} (Chegini & Ghobadion, 2005) or from 0.41 to 0.56 g cm^{-3} (Goula & Adamopoulos, 2010). However, all of the cited studies were performed on a laboratory scale, which creates different conditions for water evaporation due to varying chamber geometry and size. Powders from the chamber (Ch) were characterized by a higher loose bulk density, compared to those obtained from the cyclone container (C), significant differences were observed in the case of powders M and H25. The same dependence (a higher loose bulk density of powders from the chamber and a lower one for the powders from the cyclone container) was noted for other flavours obtained both in the laboratory and at the production scale (Jedlińska *et al*., 2013; Jedlińska *et al*., 2014). The higher loose bulk density of powders from the chamber was probably associated with a large dispersion of particle size, which is confirmed by the particle-size distribution curves – they were "spread" and shifted to the right – towards larger diameters. Smaller particles occupy spaces between the larger ones, which leads to a smaller bed volume, and thus a higher loose density. On the other

Table 2. The physical properties of microencapsulated orange aromas obtained by spray drying: after mixing from the cyclone container (MC) and from the chamber (MCh); after mixing and homogenization at 17 MPa from the cyclone container (H17C) and from the chamber (H17Ch); after mixing and homogenization at 25 MPa from the cyclone container (H25C) and from the chamber (H25Ch)

Variants	Water content WC (%)	Loose bulk density ρ_L (g cm^{-3})	Tapped bulk density ρ_T (g cm^{-3})	Particle density ρ_P (g cm^{-3})	Loose porosity ε_L (%)	Tapped porosity ε_T (%)	Hausner ratio HR	T_{g1}	T_{g2}
MC	4.5±0.0^{b}	0.39±0.01^{ab}	0.58±0.02^{b}	1.44±0.03^{a}	72.6±0.4^{bc}	58.5±0.3^{ab}	1.51±0.01^{c}	89.2±0.5	82.1±0.5^{a*}
MCh	3.5±0.9^{ab}	0.43±0.01^{c}	0.59±0.02^{b}	1.40±0.01^{a}	69.3±0.6^{a}	57.1±0.6^{a}	1.40±0.01^{b}	90.0±1.2	84.7±2.0^{a*}
H17C	3.8±0.5^{ab}	0.39±0.02^{ab}	0.53±0.01^{a}	1.33±0.03^{a}	71.6±0.5^{b}	60.3±0.5^{c}	1.39±0.01^{b}	90.1±0.6	82.9±1.1^{a*}
H17Ch	2.6±0.0^{a}	0.40±0.01^{abc}	0.52±0.01^{a}	1.29±0.01^{a}	69.0±0.5^{a}	59.7±0.5^{bc}	1.30±0.01^{a}	88.6±1.9	81.3±0.9^{a*}
H25C	4.2±0.2^{ab}	0.39±0.00^{a}	0.57±0.02^{b}	1.46±0.02^{a}	73.3±0.4^{c}	60.9±0.8^{c}	1.46±0.05^{c}	90.0±0.3	81.9±0.4^{a*}
H25Ch	2.8±0.0^{a}	0.42±0.01^{bc}	0.57±0.02^{b}	1.35±0.02^{a}	68.8±0.3^{a}	57.8±0.8^{a}	1.36±0.02^{ab}	89.1±0.5	83.0±1.0^{a*}

hand, Goula and Adamopoulos (2010), Janiszewska *et al.* (2008) and Samborska *et al.* (2015) found that loose bulk density is closely related to the water content of the powders. A higher water content causes particles to combine into larger aggregations, resulting in empty voids between them. This, in turn, causes a reduction in bulk density. The results obtained support this conclusion – powders collected from the cyclone container were characterized by a higher water content and lower loose bulk density. There was no significant effect of the method of preparing the solution on loose bulk powder density.

The particle density (ρ_P) of the powders ranged from 1.29±0.01 to 1.46±0.02 g cm^{-3} (Table 2). Similar values of apparent density, ranging from 1.22±0.1 to 1.44±7.1 g cm^{-3}, were obtained by Janiszewska *et al*. (2010), who spray dried lemon aroma solutions with a 30% concentration of the carriers (26% maltodextrin and 4% gum) and various concentrations of the flavours (2, 4, 6, 8 or 10%). The particle density of the microcapsules collected from the cyclone container was higher than it was from the chamber for each pretreatment method, but the differences were statistically insignificant. Janiszewska *et al*. (2011) noted significant differences in the particle density of the powders depending on the homogenization method, the lowest density was observed for the powders after mixing, and the highest after a two-stage pressure homogenization. However, it should be noted that these tests were carried out on a laboratory scale, not under industrial conditions.

The porosity of the loose and tapped bed of the industrial orange flavours ranged from 68.8±0.3 to 73.3±0.4% and from 57.1±0.6 to 60.9±0.8%, respectively (Table 2). It was found that the powders from the cyclone container were characterized by a significantly higher bed porosity (both loose and tapped) compared to those from the chamber. As described above, the particle-size distribution of chamber powders was more extensive, which introduced the possibility of smaller particles filling the gaps between the bigger particles, and, as a consequence decreasing bed porosity. Compared to the loose bed porosity, taking into account the same reception location, there was no significant difference between the powders obtained from emulsions treated with various pre-treatments. In the case of the tapped bed, a greater differentiation in the effect of the pre-treatment of the solution on the porosity could be observed. In the case of C powders (powders from the cyclone container) a significantly lower porosity was noted than for M powders (after mixing pre-treatment).

The Hausner ratio (HR) provides information about the cohesion of the powder related to the cohesion forces. Powders with a HR value greater than 1.4 have a high degree of cohesiveness and form a coherent structure. Good flowability, and thus low cohesiveness is typical for powders with a HR value lower than 1.2 (Hausner, 1967).

The HR of microencapsulated orange aromas varied from 1.30±0.01 to 1.51±0.01 (Table 2), indicating the high degree of cohesiveness of the powders and a coherent structure. Powders taken from the cyclone container were characterized by a greater degree of cohesion than those from the chamber, which probably resulted from the particle size – the powders from the cyclone container were characterized by smaller particle diameters than the powders from the chamber. Previous studies at the laboratory scale (Jedlińska *et al*., 2013) also confirmed that the powders taken from the cyclone container showed a significantly greater cohesion compared to those from the chamber.

By comparison, spray-dried tamarind pulp was characterized by HR values from 1.27 to 1.52 (Bhusari *et al*., 2014). It is noteworthy that there was no significant correlation between water content and the Hausner ratio ($r = 0.20$; p-value = 0.39). Powders H17Ch and H25Ch had significantly better flow properties than other samples, which may serve to prove the purposefulness of homogenization pretreatment.

The glass transition temperature T_g of the orange flavours microencapsulated by spray drying was in the range from 81.3±0.9 to 90.1±0.6°C (Table 2), so it was at a level typical for amorphous carbohydrates and sugars (Roos & Karel, 1991a; Khalloufi *et al*., 2000). Islam *et al*., (2016) presented a similar T_g value (86.63±0.97°C) for the spray dried orange juice powder with the addition of maltodextrin as a carrier. T_{g1} was significantly higher than T_{g2}, which could be a result of the fact that during the first circulation there was probably a breakdown of the maltodextrin nets and the formation of compounds with a lower molecular weight and lower glass transition temperature. The powders were stored in an environment of variable humidity (under conditions determined by the manufacturer – darkened room, room temperature, in packaging that did not guarantee full water vapour impermeability). This could have an effect on T_{g1}, so it was reasonable, during comparisons between variables, to compare Tg values taking into account T_{g2}. T_{g2} was not significantly affected by the powder collection location and the kind of pre-treatment implemented before drying. Based on previous studies, the creation of different spatial forms of chemical compounds in powders from the cyclone and the chamber could be expected due to long-term exposure to high temperatures in the chamber (the phenomenon of polymorphism), (Jedlinska *et al*., 2014), which could affect the T_g values. However, this phenomenon was not observed, the T_{g2} values were at the same level for every sample.

A qualified team from the "Pollena-Aroma" company carried out the sensory evaluation of the microencapsulated aromas obtained using the triangular type differential method: two samples the same, one different. There was no difference in the taste and smell of the powders obtained after various emulsion preparation pre-treatments. Thus, the powders obtained from the emulsions after pressure homogenization were treated as flavours obtained in regular production (only after mechanical mixing), and sent to the customers. Unfortunately, contrary to predictions, pressure homogenization, as a way of preparing emulsions for spray drying did not reduce the dosage of the aroma in the finished product.

PCA was applied to the data matrix of physicochemical properties of orange microencapsulated flavours. It was found that PC1 and PC2 explained up to 98.63% of the total variance in the physicochemical properties of orange microencapsulated flavours, 89.55% was explained by PC1 and 9.08% by PC2. In the PCA score plot (Fig. 4), almost all variables were near the circle, which means that most of the information contained in these variables was carried by the main components.

A strong positive correlation between most of the powder properties (the angle between the vectors was equal or near 0°) such as myrcene, linalyl acetate and limonene content, water content (WC), flowability (HR ratio), glass

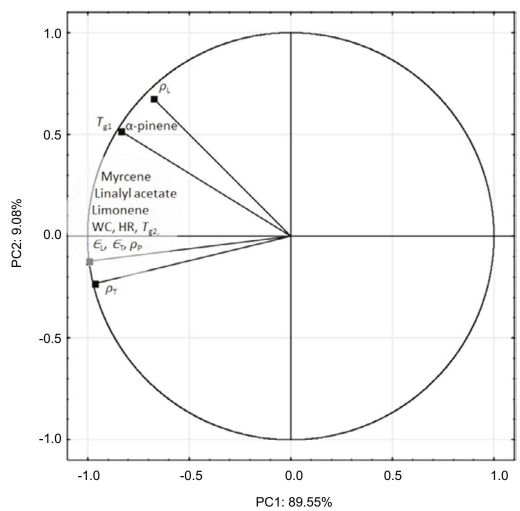

Fig. 4. Score plot.

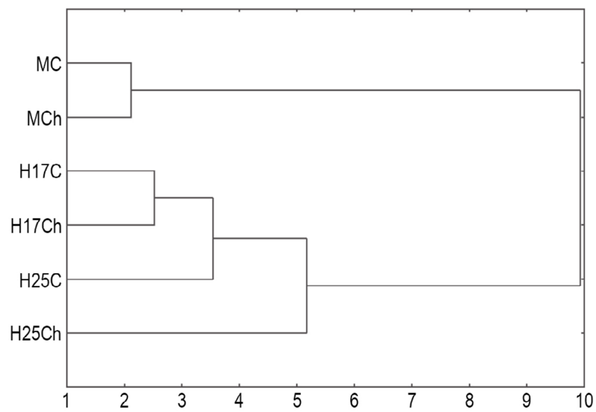

Fig. 5. Dendrogram.

temperature of the second circulation (T_{g2}), loose and tapped porosity ($\in_L$, $\in_T$), particle and tapped density (ρ_p, ρ_T) was found. The glass temperature of the first circulation (T_{g1}), α-pinene content and loose density (ρ_l) were positively correlated but less correlated than the other properties.

HCA is a clustering method, which presents the organization of samples in groups and their hierarchy (Lee and Yang, 2009; Granto *et al*., 2018). The result of HCA was presented in the dendrogram figure (Fig. 5). Two clusters were separated: cluster I containing powders M (from feed solutions after mixing), and cluster II containing powders H (from feed solutions after homogenization). The shorter distance was measured in the case of pairs MC and MCh and also H17C and H17Ch – despite the different reception locations, the powders had many common properties. The occurrence of separate clusters for the M and H samples indicates that, in the overall description there are significant differences between the powders obtained after the two pretreatment methods, this result may encourage further research.

CONCLUSIONS

1. The powders analysed for the article, were produced by the "Pollena-Aroma" company, and contained a dozen or even several dozen different compounds. The industry flavour compositions consisted of a large number of ingredients, because they should provide a specific, sophisticated fragrance, which is often difficult to imitate and stands out from the competition (clearly associated with a specific product).

2. The powders obtained from the solutions after homogenization at 17 MPa, compared to the powders from mixed solutions, some improved physical properties (better flowability, lower water content). The powders obtained from the solutions after homogenization at 25 MPa had better α-pinene and myrcene retention than the powders produced from mixed solutions. Cluster analysis showed the overall differences between the powders obtained from mixed or homogenized solutions, which may encourage further research

3. Significant differences between the powders collected from the cyclone container and the chamber of the spray dryer were found in terms of water content, bulk density, particle-size distribution and aroma retention. The powders obtained from the chamber were characterized by a lower water content and porosity, and better flowability.

Conflict of interest: The authors declare that they have no conflict of interest.

Compliance with ethical requirements: This study does not contain any experiment involving human or animal subjects.

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