Available online at www.sciencedirect.com
ScienceDirect
Food Chemistry 109 (2008) 17–24
Food Chemistry
www.elsevier.com/locate/foodchem

Extractable oil in microcapsules prepared by spray-drying: Localisation, determination and impact on oxidative stability

S. Drusch *, S. Berg

Institute of Human Nutrition and Food Science, University of Kiel, Heinrich-Hecht-Platz 10, 24118 Kiel, Germany

Received 11 September 2007; received in revised form 29 November 2007; accepted 3 December 2007

Abstract

Aim of the present study was to investigate the localisation of the extractable oil in spray-dried microencapsulated fish oil prepared under different spray-drying conditions and to investigate the impact on lipid oxidation upon storage. Confocal laser scanning microscopy, scanning electron microscopy and different extraction procedures revealed that the extractable oil in microencapsulated fish oil is mainly located on the surface and in oil droplets close to the surface. Consequently, different methods for determination of the different fractions are proposed. Lipid oxidation as determined by hydroperoxide content or anisidine value was higher in microcapsules with 50% oil load spray-dried at 210/90 °C, propanal content was increased in samples with 30% oil load spray-dried at 210/90 °C. The differences in stability could only partly be explained by the varying amount of extractable oil. It is concluded that the surface oil protects other fractions of the extractable oil and that the extractable oil cannot be used to predict shelf-life of microencapsulated oils.

Keywords: Encapsulation; CLSM; Surface oil; Non-encapsulated oil; Long chain polyunsaturated fatty acids; Lipid oxidation

1. Introduction

Microencapsulation techniques offer the possibility for the protection and controlled release of food ingredients. Generally, encapsulation techniques can be divided into three classes: chemical processes like molecular inclusion or interfacial polymerization, physicochemical techniques like coacervation and liposome encapsulation and physical processes like spray-drying, spray chilling/cooling, co-crystallization, extrusion or fluidized bed coating (Kunz, Krückeberg, & Weißbrodt, 2003). The principal technologies used for encapsulation of lipophilic food ingredients are spray-drying, coacervation and extrusion.

In spray-dried emulsions, the amount of non-encapsulated oil is a key parameter determining the product quality. It has been shown, that in fat-containing dairy powders the surface is almost completely covered with a thin layer of fat (Kim, Chen, & Pearce, 2002) and it is a well-established fact that this surface fat determines the flowability and wettability of spray-dried dairy powders (Vega & Roos, 2006). Furthermore, non-encapsulated milk fat undergoes oxidation and may lead to off-flavour formation. It has already been shown in the early 1970s for spray-dried dairy products by Buma (1971a), that the extractable oil consists of different fractions comprising (1) the surface fat, (2) the outer layer fat in the surface layer of the particle, (3) fat, that can be extracted by the solvent through capillary forces and (4) fat, that can be reached by solvent through holes left by already extracted fat (Fig. 1). Several different methods for the determination of free or surface fat in spray-dried emulsions exist (Vega & Roos, 2006) and these different methods extract different fractions of the non-encapsulated oil.

Apart from spray-drying of dairy products, it is nowadays a common practice to encapsulate sensitive ingredients like essential oils and nutritional oils rich in polyunsaturated fatty acids by spray-drying. In these applications non-encapsulated core material is particularly

* Corresponding author. Tel.: +49 431 8802370; fax: +49 431 8805544.
E-mail address: sdrusch@foodtech.uni-kiel.de (S. Drusch).

0308-8146/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.foodchem.2007.12.016

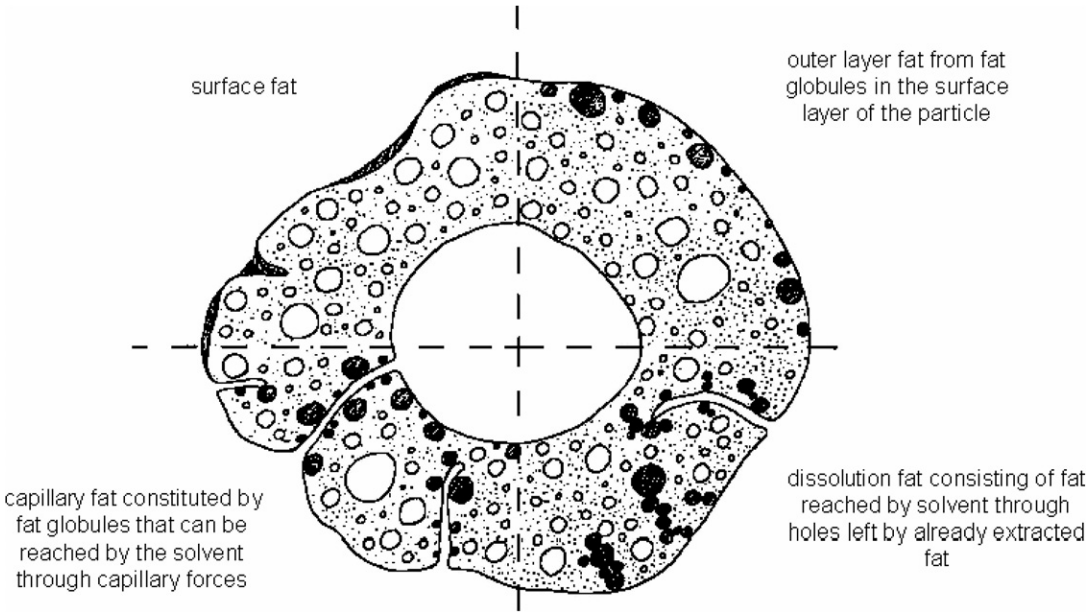

Fig. 1. Schematic model of four forms of extractable fat in a milk powder particle with a central vacuole. (Buma (1971c). Reproduced with permission from Elsevier).

prone to oxidation (Baik et al., 2004; Ponginebbi, Nawar, & Chinachoti, 1999), but it is not necessarily correlated with shelf-life of the microencapsulated product as it was shown for orange oil (Finney, Buffo, & Reineccius, 2002) or methyl linoleate (Minemoto, Adachi, & Matsuno, 1997). The latter led some authors to the misleading conclusion that non-encapsulated core material is not important in terms of product stability or shelf-life, but irrespectively of its amount, in applications like encapsulation of oils rich in polyunsaturated fatty acids non-encapsulated core material may strongly impair product acceptability. Autoxidation of polyunsaturated fatty acids leads to the development of volatile secondary oxidation products. In terms of sensory perception of oxidised fish oil 1-penten-3-one, (*Z*)-4-heptanal, 1-octen-3-one (*Z*)-1,5-octadien-3-one, (*E*,*E*)-2,4-heptadienal and (*E*,*Z*)-2,6 nonadienal have been identified as important volatiles (Venkateshwarlu, Let, Meyer, & Jacobsen, 2004). Generally, the secondary lipid oxidation products may have a very low odour threshold and consequently negatively affect sensory of the microcapsules (Jacobsen, 1999; Lee et al., 2003; Venkateshwarlu et al., 2004) and limit the shelf-life of these products.

Aim of the present study was to investigate the localisation of the extractable oil in spray-dried microencapsulated fish oil prepared under different spray-drying conditions and to investigate the impact on lipid oxidation upon storage.

2. Materials and methods

Fish oil (Fish oil 18/12, Cognis Deutschland GmbH & Co. KG, Illertissen, Germany) was microencapsulated at two different oil loadings, 30% and 50% into a matrix of *n*-octenylsuccinate-derivatised starch (nOSA starch; HiCap100, National Starch & Chemicals GmbH, Hamburg, Germany) and glucose syrup (C*Dry 01934, Cargill Deutschland GmbH, Krefeld, Germany). Parent emulsions were homogenised at 500/100 bar with two passes (Panda 2K, Niro Soavi Deutschland, Lübeck, Germany) and subsequent spray-drying (Mobile Minor, Niro A/S, Denmark) was performed at two different temperature settings of 160/60 °C and 210/90 °C. Process variation and two different oil loads were included to induce changes in particle morphology, thus providing samples with a different proportion of surface oil and inner non-encapsulated oil.

Total oil content of the microcapsules was analysed following the Mojonnier method (BVL L 02.06-E(EG) and 1(EG) to 8(EG):1981-01, Method 4 Appendix II: Analytical method regarding the composition of certain partially or completely dried non-perishable milk products). Briefly, the sample is hydrolyzed with ammonia and the oil is extracted with ethanol, diethyl ether and petrol ether. After removal of the organic solvent the oil content is determined gravimetrically. The determination of non-encapsulated oil was performed using three different methods. The first method was based on a Soxhlet extraction with petrol ether as organic solvent. It is the basis for the officially recommended method for fat determination in a variety of food products, e.g. the determination of free fat in dry milk products (VDLUFA, 1995). The extraction was performed with 7, 15 or 24 passes of the solvent through the sample. The second method for determination of the non-encapsulated oil is a rapid method described by Westergaard (2004). Briefly, 10 g of sample are weighed into a screw-capped flask. Petrol ether (50 ml) is added and the sample

is mixed slowly for 15 min. The dispersion is filtered and 25 ml of the filtrate are evaporated to dryness. The residual oil is weighed and the percentage of extractable oil is calculated. The third method for determination of the extractable oil was a two-step extraction according to Kim, Chen, and Pearce (2005). To extract the surface oil from the microcapsules, 2 g of powder were placed on a filter and washed with 4 × 10 ml of hexane. Inner fractions of non-encapsulated oil were subsequently extracted from the same powder using 1 g of the powder and 40 ml hexane, the extraction time was 48 h.

For visualisation of the oil in the microcapsules, the fluorescent dye nile red was dissolved in the oil phase prior to spray-drying. Confocal laser scanning microscopy (CLSM) was performed using a Leica TCS SP (Leica-Microsystems, Wetzlar, Germany). Samples were analysed using a He and a Ne–Ar-laser operated at 488 nm. All fluorescent pictures were taken using an 63 × oil immersion objective. The software for CLSM imaging was LCS Pro (Leica-Microsystems, Wetzlar, Germany). A CamScan 44 REM/EDX scanning electron microscope (CamScan USA Inc, Cranberry Township PA, USA) was used to view the characteristics of the microcapsules.

The different microcapsules were stored at 33% relative humidity at 20 °C for a period of eight weeks. Lipid oxidation was monitored through the analysis of hydroperoxide content, the anisidine value and propanal, a major volatile secondary lipid oxidation product, by static headspace gas chromatography with mass selective detection. Hydroperoxide content was determined using the IDF standard method 74A:1991 for the determination of the peroxide value in anhydrous milk fat with slight modifications (International Dairy Federation, 1991). 2-Propanol was used as solvent in the test protocol. After addition of the iron-II-chloride and the ammonium thiocyanate solution, samples were incubated in a water bath at 60 °C for 30 min. The extinction was measured at a wavelength of 485 nm. The anisidine value was determined as described in the official method for determination of the anisidine value in fats and oils (BVL L 13.00-15:2001-07 Untersuchung von Lebensmitteln – Tierische und pflanzliche Fette und Öle – Bestimmung der Anisidinzahl (Übernahme der gleichnamigen DIN EN ISO 6885, Ausgabe Dezember 2000). Propanal was analysed via static headspace chromatography with mass selective detection as described by Drusch, Serfert, Scampicchio, Schmidt-Hansberg, and Schwarz (2007).

Statistical analysis was performed using Design Expert, Version 6.0.10 (Stat Ease Inc, Minneapolis, USA). For the data on lipid oxidation a two-level full-factorial design with two replicates and the categorical factors "oil load" and "drying conditions" was created. Hydroperoxide content, anisidine value and proapanal content at each day of analysis were chosen as response. Within this design, power at 5% alpha level for effect of twofold standard deviation was 57.2%. Probability values from the ANOVA are reported.

3. Results and discussion

Composition and spray-drying conditions may affect the amount and the distribution of non-encapsulated oil. In the present study process variation and two different oil loads were included to induce changes in particle morphology, thus providing samples with a different proportion of surface oil and inner non-encapsulated oil. Extractable oil determined by the extraction procedure of Kim et al. (2005) ranged from 1.83 to 13.5% for the microcapsules with 30% oil dried at 160/60 °C and the microcapsules with 50% oil load dried at 210/90 °C, respectively (Table 1).

As reviewed by Vignolles, Jeantet, Lopez, and Schuck (2007), free fat content in dairy powders is linearly correlated with total fat content and the authors cite Young, Sarda, and Rosenberg (1993), who explained this correlation with a decreased thickness of the encapsulate around the fat droplets. In the present study with 0.84 μm the oil droplet size was lower in the parent emulsion with 30% oil load compared to 1.29 μm in the parent emulsion with 50% oil load (Table 2). The increase in oil droplet size may partly explain the higher amount of non-encapsulated oil in the samples with 50% oil load, but cannot explain the differences between the two drying conditions at a fixed oil load. Soottitantawat, Yoshii, Furuta, Ohkawara, and Linko (2003) report a lower retention of orange oil during encapsulation and a higher amount of surface oil at increased oil droplet size in the parent emulsion. The authors speculate that shearing and rupture of large oil droplets during atomization may be responsible for these effects. In the present study, an increase in oil droplet size has been observed after the drying process and thus, the fraction of non-encapsulated oil partly arises from oil droplet coalescence during the drying process. This has already been observed in previous trials, in which non-encapsulated oil amounted up to 25% (Drusch, Serfert, Scampicchio, Schmidt-Hansberg, & Schwarz, 2007).

The difference in extractable oil caused by different drying conditions may be attributed to both a higher inlet and outlet temperature at 210/90 °C. For both parameters an increase in extractable fat content has bee reported due to formation of vacuoles and pores (Sloth Hansen, 1980) or formation of cracks (Bhandari, Dumoulin, Richard, Noleau, & Lebert, 1992). Furthermore the difference in particle size accounts for the increase in extractable oil at different drying conditions. Generally large particles have a lower surface to volume ratio and thus can encapsulate more oil. This has been shown, e.g. for dairy powders (Buma, 1971b) and orange oil encapsulated in a modified starch matrix (Finney et al., 2002).

In the present study irrespectively of the oil load, the amount of non-encapsulated oil extracted during the second step of the extraction according to Kim et al. (2005) was always below 1% being slightly higher in microcapsules dried at 210/90 °C. Thus, a high proportion of the non-encapsulated oil is located on the surface and in outer layer oil from oil droplets in the surface layer of the particle.

Table 1
Total oil (%) and non-encapsulated oil (% of the total oil) in microencapsulated fish oil dried at 160/60 or 210/90 °C as determined by different extraction procedures

	Total oil	Westergaard (2004)	VDLUFA (1995)	Kim et al. (2005), 1st extraction	Kim et al. (2005), 2nd extraction	Kim et al. (2005), total
30% oil, 160/60 °C	30.7 ± 0.02	0.99 ± 0.03	0.92 ± 0.29	1.33 ± 0.49	0.51 ± 0.16	1.83
30% oil, 210/90 °C	31.1 ± 0.03	2.44 ± 0.03	1.98 ± 0.71	2.51 ± 0.19	0.8 ± 0.38	3.31
50% oil, 160/60 °C	51.0 ± 0.08	6.33 ± 0.13	4.58 ± 0.43	6.44 ± 0.63	0.39 ± 0.27	6.84
50% oil, 210/90 °C	49.1 ± 0.01	11.3 ± 0.17	9.03 ± 1.12	12.7 ± 0.81	0.78 ± 0.48	13.5

Table 2
Particle size and oil droplet size of microencapsulated fish oil dried at 160/60 or 210/90 °C

Sample	Particle size		Oil droplet size, parent emulsion		Oil droplet size, reconstituted emulsion	
	50 percentile (μm)	90 percentile (μm)	50 percentile (μm)	90 percentile (μm)	50 percentile (μm)	90 percentile (μm)
30% oil, 160/60 °C	21.6 ± 0.44	46.5 ± 1.35	0.84 ± 0.00	1.57 ± 0.01	0.92 ± 0.00	1.78 ± 0.00
30% oil, 210/90 °C	16.6 ± 0.16	37.9 ± 2.63	0.84 ± 0.00	1.57 ± 0.01	0.99 ± 0.02	2.20 ± 0.16
50% oil, 160/60 °C	18.9 ± 0.04	45.5 ± 1.85	1.29 ± 0.03	3.38 ± 0.08	2.37 ± 0.13	6.57 ± 0.41
50% oil, 210/90 °C	15.8 ± 0.33	35.3 ± 3.11	1.29 ± 0.03	3.38 ± 0.08	4.24 ± 0.09	14.1 ± 0.15

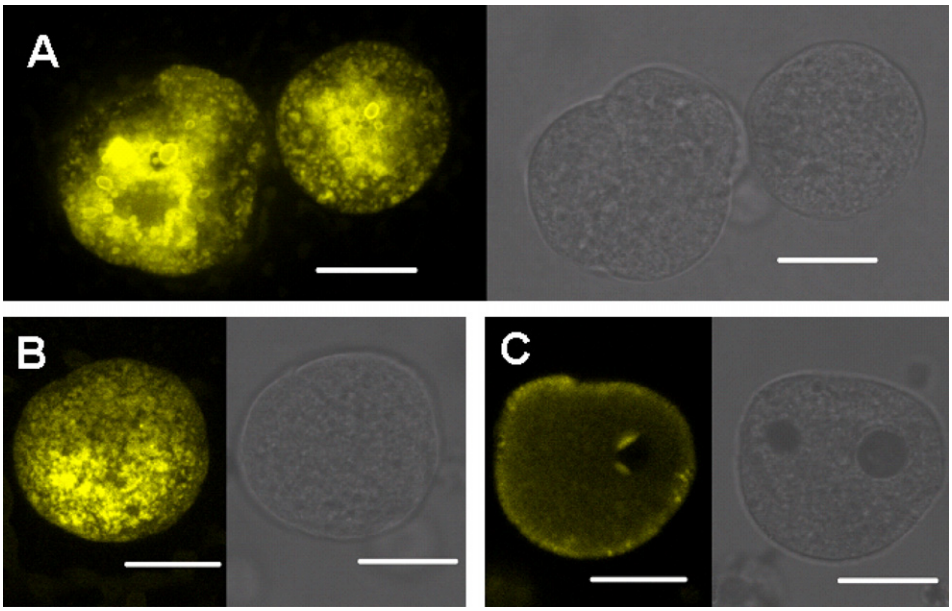

Fig. 2. CLSM images of nOSA-starch-based microcapsules containing 50% oil (A) without extraction, (B) after extraction of the surface oil and (C) the inner free oil according to Kim et al. (2005).

CLSM showed a high proportion of the non-encapsulated oil on the powder surface (Fig. 2A) and scanning electron microscopy showed a higher proportion of oil droplets on the surface of microcapsules with 50% oil load compared to microcapsules with 30% oil load (Fig. 3).

These results are in agreement with the observations of Kim et al. (2002), who showed using electron spectroscopy for chemical analysis (ESCA) that in whole milk powder fat covers the whole spray-dried particle. However, ESCA only analyses the outermost surface layer of approximately 10 nm (Kim et al., 2005) and is thus not suitable for quantifying surface oil (Vega & Roos, 2006). In the present study, after extraction of the surface oil, a micro- and mesoporous structure of the surface appeared and intensity of fluorescence, which is now only attributed to pores filled with oil, decreased (Fig. 2B).

The extraction procedure of Westergaard (2004) showed similar results compared with the first extraction of Kim et al. (2005) and thus is suitable for determination of the surface oil (Table 1). In microcapsules with 50% oil load, in which this fraction represents nearly all of the non-encapsulated oil, the method of Westergaard (2004) may be used for an approximation of the non-encapsulated oil. In contrast, in samples with a low proportion of non-encapsulated oil, in which surface oil only represents a low proportion of the non-encapsulated oil, the method of Westergaard (2004) may severely underestimate the fraction of non-encapsulated oil as determined by Kim et al.

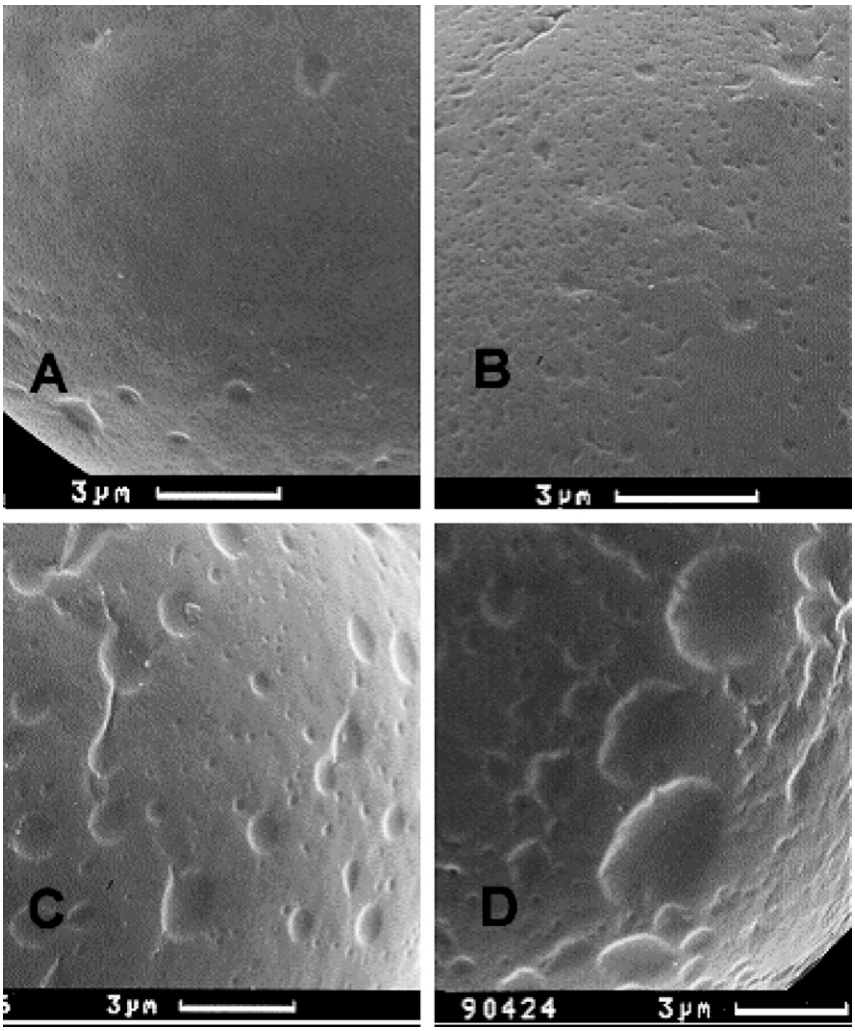

Fig. 3. Scanning electron micrographs of the particle surface of microencapsulated fish oil with (A, B) 30% or (C, D) 50% oil load spray-dried at 160/60 °C (A,C) or 210/90 °C (B, D).

(2005). Following a Soxhlet extraction procedure with seven passes of the solvent through the sample (VDLUFA, 1995) comparably low amounts of non-encapsulated oil were extracted. In another experiment, it was shown that increasing the number of passes up to 24 passes of the solvent through the samples steadily increases the amount of extracted oil (data not shown).

Lipid oxidation of microencapsulated oils is affected by microcapsules characteristics. Generally, at elevated inlet air temperature particle ballooning may occur (Finney et al., 2002; Walton & Mumford, 1999). Drusch and Schwarz (2006) investigated the effect of spray-drying conditions on particle characteristics and oxidation of microencapsulated fish oil and showed that high inlet and outlet air temperature may lead to ballooning and autoxidation of the encapsulated and non-encapsulated core material already during the drying process, if a wall material with strong film forming properties is used. In the present study, a nOSA-starch with a high and precisely defined degree of hydrolysation, mainly consisting of disaccharides, has been chosen and thus, based on particle size analysis, no ballooning was observed at high drying temperature of 210/90 °C (Table 2).

Upon storage the hydroperoxide content in all samples steadily increased (Fig. 4). The increase in peroxide value was significantly higher in the sample with 50% oil load spray-dried at 210/90 °C compared to all other samples ($p < 0.01$). After eight weeks of storage hydroperoxide content amounted to 211 ± 6.9 mmol/kg oil in this sample, whereas it only increased up to 141 ± 5.7 mmol/kg oil in all other samples. The anisidine value also showed a steady increase over the storage period. From day 28 on the anisidine value was significantly increased in the sample with 50% oil load dried at 210/90 °C ($p < 0.01$). After 56 days of storage peroxide value and anisidine content were significantly increased in samples with 50% oil load compared to samples with 30% oil load ($p < 0.05$). Generally, hydroperoxide content and anisidine value showed a good correlation ($r = 0.941$), but higher values for the sample with 50% oil load dried at 210/90 °C were first observed after

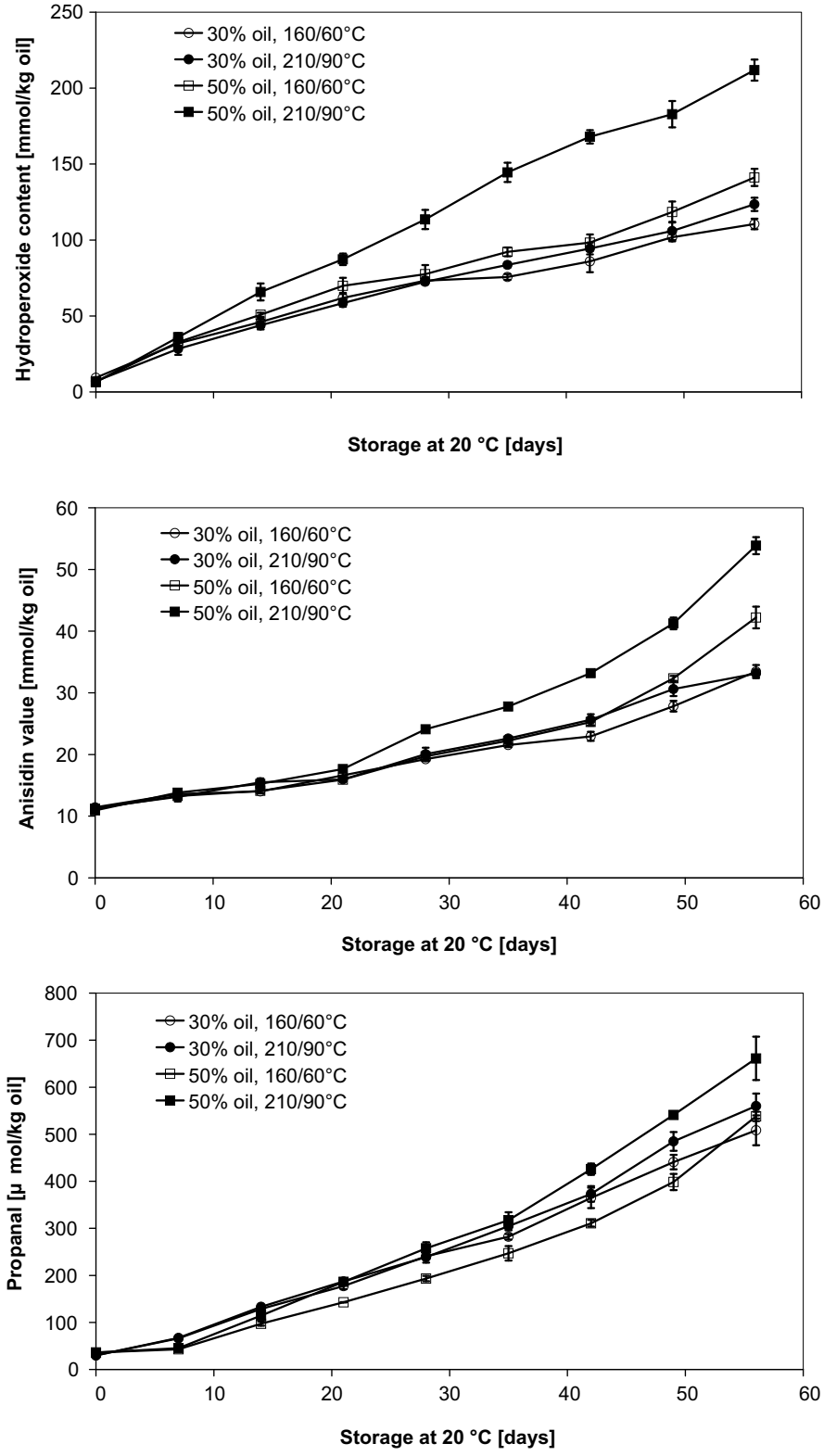

Fig. 4. Development of hydroperoxide content, anisidine value and propanal content in microencapsulated fish oil stored for eight weeks at 20 °C and 33% relative humidity.

three weeks of storage. With $r = 0.906$ and $r = 0.956$, the development of propanal correlated well with hydroperoxide content and anisidine value. Extractable oil was significantly influenced by the oil load and, at a high oil load, also by drying conditions ($p < 0.05$). Therefore the course of lipid oxidation cannot solely be explained by the amount

of extractable oil. Keogh et al. (2001) showed, that air inclusion influences the shelf-life of microencapsulated fish oil. As discussed above, in the present study particle ballooning did not occur and since, the samples were prepared from the same parent emulsion, differences in air inclusion caused by discrete inclusion of air in the parent emulsion and subsequently in the microcapsules can be excluded. Drusch et al. (2007) showed that micro-structural particle characteristics related to the type of emulsifier and particle characteristics related to the viscosity of the parent emulsion and its drying behaviour are major determinants of the microcapsule stability.

A comparison of the course of lipid oxidation with data reported in other encapsulation studies for oils rich in long chain polyunsaturated fatty acids is generally difficult due to differences in the core material stabilization, the wall material used for encapsulation, the process and storage conditions and the parameters used to monitor lipid oxidation.

In conclusion, non-encapsulated oil in microencapsulated fish oil is mainly located on the surface and in oil droplets close to the surface. Thus, following the extraction procedure described by Westergaard (2004) or by Kim et al. (2005), the amount of surface oil in microcapsules prepared by spray-drying can be determined. For complete extraction of the non-encapsulated oil the extraction procedure of Kim et al. (2005) or Soxhlet extraction with an adequate number of passes of solvent ($n > 24$) must be used. Lipid oxidation as determined by hydroperoxide content or anisidine value was higher in microcapsules with 50% oil load spray-dried at 210/90 °C, propanal content was increased in samples with 30% oil load spray-dried at 210/90 °C. The differences in stability could only partly be explained by the varying amount of extractable oil. It is concluded that the surface oil protects other fractions of the extractable oil and that the extractable oil cannot be used to predict shelf-life of microencapsulated oils.

Acknowledgements

The present work was funded by the Arbeitsgemeinschaft industrieller Forschungsvereinigungen "Otto von Guericke" e.V. (AiF 14951N). Scanning electron microscopy was performed at the Institut für Geowissenschaften, Abt. Geologie, University of Kiel, confocal laser scanning microscopy at the Zentrale Mikroskopie, University of Kiel. The authors gratefully acknowledge the skilful help of Ute Schuldt, Maria Mulisch and Jörg Knipp.

References

Baik, M.-Y., Suhendro, E. L., Nawar, W. W., McClements, D. J., Decker, E. A., & Chinachoti, P. (2004). Effects of antioxidants and humidity on the oxidative stability of microencapsulated fish oil. *Journal of the American Oil Chemists Society, 81*(4), 355–360.

Bhandari, B. R., Dumoulin, E. D., Richard, H. M. J., Noleau, R. I., & Lebert, A. M. (1992). Flavor encapsulation by spray-drying: Application to citral and linalyl acetate. *Journal of Food Science, 57*(1), 217–221.

Buma, T. J. (1971a). Free fat in spray-dried whole milk. 1. General introduction and brief review of literature. *Netherlands Milk and Dairy Journal, 25*, 33–41.

Buma, T. J. (1971b). Free fat in spray-dried whole milk. 3. Particle size. Its estimation, influence of processing parameters and its relation to free-fat content. *Netherlands Milk and Dairy Journal, 25*, 53–72.

Buma, T. J. (1971c). Free fat in spray-dried whole milk. 10. A final report with a physical model for free fat in spray-dried milk. *Netherlands Milk and Dairy Journal, 25*, 159–174.

Drusch, S., & Schwarz, K. (2006). Microencapsulation properties of two different types of *n*-octenylsuccinate-derivatised starch. *European Food Research and Technology, 222*, 155–164.

Drusch, S., Serfert, Y., Scampicchio, M., Schmidt-Hansberg, B., & Schwarz, K. (2007). Impact of physicochemical characteristics on the oxidative stability of fish oil microencapsulated by spray-drying. *Journal of Agricultural and Food Chemistry, 55*(26), 11044–11051.

Finney, J., Buffo, R., & Reineccius, G. A. (2002). Effects of type of atomization and processing temperature on the physical properties and stability of spray-dried flavours. *Journal of Food Science, 67*(3), 1108–1114.

International Dairy Federation. (1991). Anhydrous milk fat. Determination of peroxide value. International IDF Standards, Square Vergot 41, Brussels, Belgium, sec. 74A:1991.

Jacobsen, C. (1999). Sensory impact of lipid oxidation in complex food systems. *Fett/Lipid, 101*, 484–492.

Keogh, M. K., O'Kennedy, B. T., Kelly, J., Auty, M. A., Kelly, P. M., Fureby, A., & Haahr, A.-M. (2001). Stability to oxidation of spray-dried fish oil powder microencapsulated using milk ingredients. *Journal of Food Science, 66*(2), 217–224.

Kim, E. H.-J., Chen, X. D., & Pearce, D. (2002). Surface characterization of four industrial spray-dried dairy powders in relation to chemical composition, structure and wetting property. *Colloids and Surfaces B: Biointerfaces, 26*, 197–212.

Kim, E. H.-J., Chen, X. D., & Pearce, D. (2005). Melting characteristics of fat present on the surface of industrial spray-dried dairy powders. *Colloids and Surfaces B: Biointerfaces, 42*, 1–8.

Kunz, B., Krückeberg, S., & Weißbrodt, J. (2003). Chancen und Grenzen der Mikroverkapselung in der modernen Lebensmittelverarbeitung. *Chemie Ingenieur Technik, 75*(11), 1733–1740.

Lee, H., Kizito, S. A., Weese, S. J., Craig-Schmidt, M. C., Lee, Y., Wei, C.-I., & An, H. (2003). Analysis of headspace volatile and oxidized volatile compounds in DHA-enriched fish oil on accelerated oxidative storage. *Journal of Food Science, 68*(7).

Minemoto, Y., Adachi, S., & Matsuno, R. (1997). Comparison of oxidation of methyl linoleate encapsulated with gum arabic by hot-air-drying and freeze-drying. *Journal of Agricultural and Food Chemistry, 45*, 4530–4534.

Ponginebbi, L., Nawar, W. W., & Chinachoti, P. (1999). Oxidation of linoleic acid in emulsions: Effect of substrate, emulsifier, and sugar concentration. *Journal of the American Oil Chemists Society, 76*, 131–138.

Sloth Hansen, P. (1980). Production of agglomerated fat-filled powder. *Journal of the Society of Dairy Technologists, 33*, 19–23.

Soottitantawat, A., Yoshii, H., Furuta, T., Ohkawara, M., & Linko, P. (2003). Microencapsulation by spray drying: Influence of emulsion size on the retention of volatile compounds. *Journal of Food Science, 68*(7), 2256–2262.

VDLUFA. (1995). Bestimmung von freiem Fett in fetthaltigen getrockneten Milcherzeugnissen. Verband der Deutschen Landwirtschaftlichen Untersuchungs- und Forschungsanstalten. Handbuch der landwirtschaftlichen Versuchs- und Untersuchungsmethodik (Methodenbuch), Band VI, C15.2.4.

Vega, C., & Roos, Y. H. (2006). Spray-dried dairy and dairy-like emulsions – compositional considerations. *Journal of Dairy Science, 89*, 383–401.

Venkateshwarlu, G., Let, M. B., Meyer, A. S., & Jacobsen, C. (2004). Chemical and olfactometric characterization of volatile flavor compounds in a fish oil enriched milk emulsion. *Journal of Agricultural and Food Chemistry, 52*, 311–317.

Vignolles, M.-L., Jeantet, R., Lopez, C., & Schuck, P. (2007). Free fat, surface fat and dairy powders: interactions between process and product. A review. *Lait, 87*, 187–236.

Walton, D. E., & Mumford, C. J. (1999). The morphology of spray-dried particles. The effect of process variables upon the morphology of spray-dried particles. *Trans IChemE, 77A*, 442–460.

Westergaard, V. (2004). Milk powder technology. In *Evaporation and spray-drying* (5th ed.) Copenhagen: Niro A/S.

Young, S. L., Sarda, X., & Rosenberg, M. (1993). Microencapsulation properties of whey proteins. 1. Microencapsulation of anhydrous milk fat. *Journal of Dairy Science, 76*, 2868–2877.