Final report of project 2819:
Emulsification of triglyceride phases in cereal matrices via high-speed extrusion process for the stabilization of lipophilic bioactive components in starch-based products

Scholar: M. Azad Emin

Supervisor: Prof. Dr. –Ing. Heike P. Schuchmann

Adress: Institute of Process Engineering in Life Sciences, Section 1: Food Process Engineering, Karlsruhe Institute of Technology, Kaiserstraße 12, 76131 Karlsruhe.

Summary: Optimizing food processes demands controlling flow of complex food matrices. CFD simulations are an important instrument in understanding it on a microscopic level, especially when measurement techniques do not really exist or cannot be applied as in processes in which high pressures, temperatures and flow velocities act on multi-phase systems. In this study, local flow of complex food matrices in an extrusion (i.e. twin screw extruder) was simulated by ANSYS POLYFLOW®. Furthermore, to investigate the dispersion of low viscous oil droplets (i.e. middle chain triglycerides) in plasticized starch matrices under extrusion-type conditions, experiments by means of a specific shear cell were performed. The efficiency of dispersive mixing of food-grade triglycerides of Newtonian behaviour in rheologically complex plasticized starch matrices was evaluated. Simulation results were validated quantitatively by experimental data obtained from extrusion trials.

Final Report:

Extrusion cooking, as a multi-unit thermal/mechanical process, has permitted a large number of food applications such as ready-to-eat cereals; salty and sweet snacks. Parallel to the increased applications, interest has grown in the physico-chemical, functional and nutritionally relevant effects of extrusion processing [1]. Prevention or reduction of nutrient destruction, together with functional fortification, is clearly of importance in most extrusion applications. Especially encapsulation of lipophilic bioactives (e.g. carotenoids, phytosterols) into extruded food matrices promises potential health benefits. However, extrusion is a complex multivariate process and presents many challenges to accomplish, particularly with respect to the stability of the bioactive lipids and the cereal matrix during processing and storage and the need to prevent undesirable interactions with the carrier food matrix.

For the encapsulation of lipophilic bioactives a lipophilic carrier is required, e.g. triglycerides. Triglyceride droplets, especially such of small sizes (e.g. < 1 µm) are known to encapsulate high concentrations of bioactive substances and improve their stability, bioavailability and palatability [2]. In laminar flow, as mainly found in extruders, shear and elongational stresses are responsible for droplet deformation and break-up as well as the molecular status of starch bio-polymers presenting the encapsulating matrix material. Hence, the knowledge of dispersion mechanisms (under elongational and shear stresses) inside the extruder is the key to control resulting emulsion characteristics, and thus bioavailability and stability of lipophilic bioactive ingredients encapsulated within the oil droplets in starch matrix. However, complex geometry of the processing rooms in extruders, especially twin-screw extruders, leads to a wide and inhomogeneous distribution of the stresses in the flow field. Therefore, measurement of the exposed local stresses in extruders is a very difficult task, if not impossible. Computational fluid dynamics (CFD) simulation of flow characteristics and mixing ability inside the extruder thus is a promising tool improving the understanding of underlying mechanisms and depicting the influence of process parameters. Furthermore, experiments under controlled stresses are crucial in order to differentiate the influence of simple shear and elongation on resulting emulsion characteristics.

In order to investigate the dispersion of medium chain triglycerides in thermoplasticized starch matrices under extrusion-type conditions, experiments by means of a specific shear cell were performed. With this device, developed at Wageningen University, native and pre-degraded starch was subjected to controlled simple shear flow. Pre-degradation was performed in a pilot plant extruder at production conditions, since these are responsible for a degradation of the bio-polymer molecules resulting in different rheological behaviour. The influence of simple shear flow (e.g. shear rate) and material characteristics (pre-degradation of starch molecules, oil content, water content, addition of emulsifier) on droplet formation, break-up and coalescence in thermoplasticized starch matrix was investigated. The shear cell has a cone/cone geometry with a constant degree of angle between the cones and therefore constant shear rates could be exerted all over the flow domain allowing to measure the viscosity of plasticized starch matrix in the same experiment. Results of these were used for analyzing droplet breakup in simple shear flow concerning critical capillary number which is depicted on Figure 1.

Furthermore, we have investigated the influence of process and material parameters (e.g. screw speed, screw design, water content, feed rate) of innovative high-speed extrusion process on dispersive and distributive mixing of triglycerides into starch based matrix by the aid of computational fluid dynamics simulation and experimental study. In order to check the physical relevance of the results obtained from numerical simulation and validate simulated data, pressure sensors were mounted on the inlet and outlet region of the mixing zone in the extruder being simulated. In order to monitor the rheological behaviour of plasticized starch matrix at different process conditions, the multiple step online slit-die rheometer was mounted on the extruder during the validation experiments. Obtained rheological data and applied process parameters (e.g. screw speed, flow rate and screw geometry) were used to conduct computational simulation of the flow in extruder parallel to the experiments. Pressure differences between inlet and outlet of the flow domain of the simulation were compared to the experimental values obtained by the inline pressure sensors.

Pressure differences obtained from computation vary depending on the time step (screw position) chosen. Similarly, the values measured during the experiments showed deviations according to technical limitations of the pressure sensors. Therefore, the mean values were calculated for both cases, and used for the validation. Results and relevant deviations are given in Table 1, which show good agreement between experimental and computational values for different process conditions.

Table 1: *Pressure difference in the simulated extruder zone: comparison of experimental and computational results for different process conditions.*

	Process condition 1	Process condition 2	Process condition 3
Flow rate (kg/h)	13	11	26
Screw speed (rpm)	200	200	400
Water content (%)	30	18	30
Viscosity ($\eta\,(\mathrm{Pa.s}) = k \cdot \dot{\gamma}^{(n-1)}$)	n = 0.15 , k = 21,583	n = 0.11 , k = 41,300	n = 0.15 , k = 21,583
Δp experimental (bar)	14,5 (± 0,2)	30 (± 0,3)	16,2 (± 0,2)
Δp computational (bar)	14,3 (± 1,5)	30,5 (± 3,2)	16 (± 1,7)

Figure 2 shows the maximum shear stress experienced by droplets during their flow in the extruder at two different screw speeds of 200 rpm and 400 rpm which were obtained from computational simulation. It is generally expected that increasing screw speed should lead to increase in maximum shear stresses since it increases the shear rate, and thus the shear stress. However, there is a negative correlation between shear stress and starch viscosity since higher shear stresses lead to higher degree of starch degradation which further results in lower viscosity, and thus to the lower maximum stresses. Maximum shear stress is directly related to viscous dissipated energy input, and thus is an important parameter on evaluating the stability of heat and/or shear labile bioactives.

Moreover, maximum capillary number experienced by the droplets in the extruder can be calculated from maximum shear stress, which can be further used to calculate maximum capillary ratio (max Ca/Ca_{crit}) with the critical capillary number (Ca_{crit}) obtained from simple shear experiments. These results were depicted on Figure 3 which shows that maximum critical capillary ratio can be increased by increasing screw speed. According to these findings, smaller droplets sizes are expected at higher screw speeds. In order to examine this suggestion, experiments at different screw speeds were performed with a twin-screw extruder (Coperion Werner & Pfleiderer, ZSK 26 Mc) which are showed in Figure 4. Obtained experimental results showed good agreement with the computational results since increasing screw speed from 400 rpm to 800 rpm led to smaller droplet sizes. Results obtained from extrusion experiment at 200 rpm are not plotted on Figure 4, since the oil drainage during the flow of samples from extrusion die, proving an inefficient dispersive mixing, was observed.

Obtained results shows the importance and necessity of parallel investigations on computational simulations and experimental studies in order to obtain reasonable evaluation of dispersive mixing of triglycerides in plasticized starch matrix in the extruder.

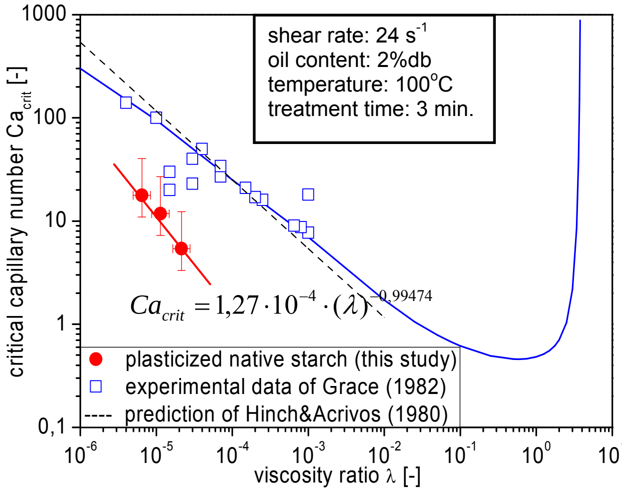

Figure 1: *Critical capillary number of triglyceride droplets in plasticized starch as a function of viscosity ratio.*

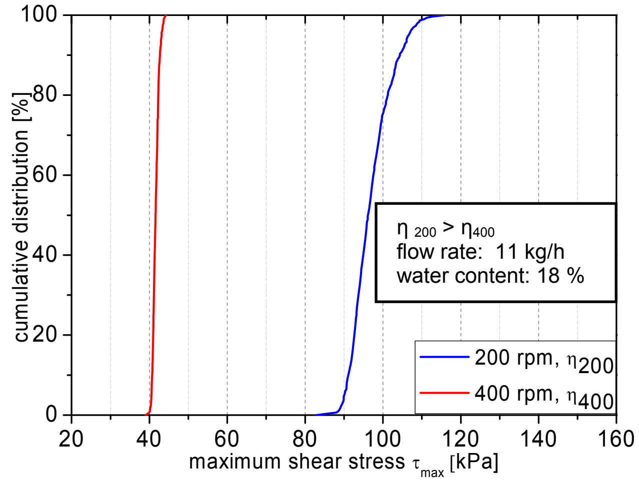

Figure 2: *Maximum shear stress experienced by droplets during their flow in the extruder at two different screw speeds of 200 rpm and 400 rpm.*

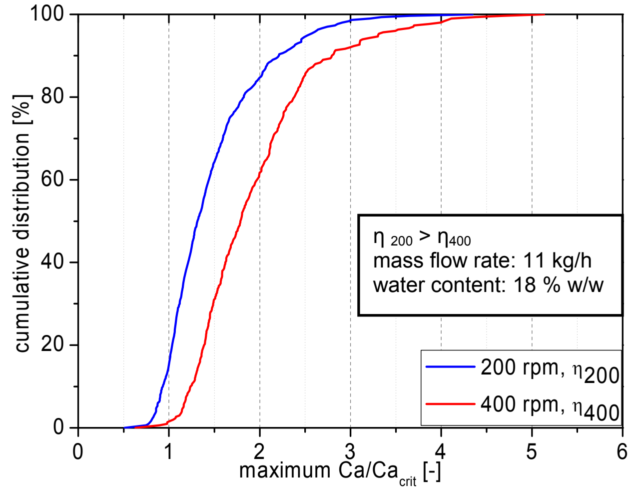

Figure 3: *Cumulative distribution of the maximum capillary ratio of the droplets at two different screw speeds of 200 rpm and 400 rpm.*

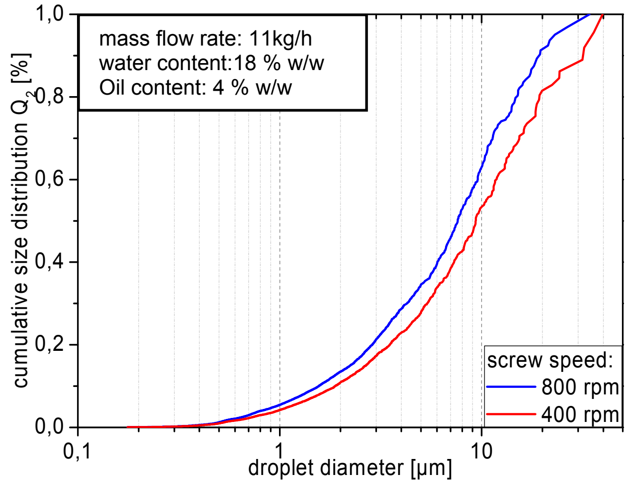

Figure 4: *Cumulative droplet size distribution obtained from extrusion experiments at two different screw speeds of 200 rpm and 400 rpm.*

References

[1] Galvin M. A., Kiely M, Flynn A. (2002). Impact of ready-to-eat breakfast cereal (RTEBC) consumption on adequacy of micronutrient intakes and compliance with dietary recommendations in Irish adults, Public Health Nutrition: 6(4), 351–363.

[2] Horn D. and Rieger J. (2001) Organic Nanoparticles in the Aqueous Phase–Theory, Experiment, and Use, Angew. Chem. Int. Ed. 2001, 40, 4330- 4361.