Thesis of Doctoral Dissertation

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MAGYAR AGRÁR- ÉS ÉLETTUDOMÁNYI EGYETEM

STUDYING LIPID-CARBOHYDRATE INTERACTIONS IN MODEL SYSTEMS

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Background to the work, objectives

Processed foods consist of numerous components that together form a multiphase system, with the main constituents being lipids, carbohydrates, and proteins. In the confectionery industry, lipids and carbohydrates have the biggest significance out of these. For food products, the study of the crystallisation phenomena of fats is essential.

The crystal structure affects the texture, quality, and sensory perception of foods. In confectionery and bakery products, fats are not present alone but are typically mixed with other components, predominantly carbohydrates. These combined components are responsible for the texture, storage-related changes, and sensory properties of products such as filling creams. Even though these two macronutrients are usually present together, there is only few research focused solely on their mixing and the interactions between them. This is why I chose to study these interactions in my doctoral research.

During my doctoral research, my objective was to investigate the structure-forming properties of lipid-carbohydrate complexes. In my experiments, I aimed to explore the effects of carbohydrates on fats. For the initial model system, I selected a mixture of a typical confectionery fat (palm mid-fraction) and a typical confectionery carbohydrate (sucrose). Subsequently, I modified both the lipid and carbohydrate phases. My aim was to map the influence of sucrose on different fats by **altering the material properties of the lipid phase**. To achieve this, I extended my research to include lauric fats and animal fats used for similar purposes as palm fat.

Furthermore, I aimed to examine the effects of various carbohydrates on palm fat-based systems. Specifically, I sought to determine how the behaviour of lipid-carbohydrate complexes changes depending on the **complexity of the carbohydrates**. That is, how mono-, di-, oligo-, and polysaccharides influence the key physical properties of palm fat-based fat creams.

Another objective of my research was to investigate how **the particle size of carbohydrates** impacts the physical properties of fat creams. To establish this, I conducted experiments using sucrose with two different particle size distributions.

To achieve the above objectives, I focused on studying the key physical parameters of the creams, including their thermal behaviour, rheology, and texture.

As a supplementary aspect of my work, I explored the potential effects of **sugar replacement** on the lipid-carbohydrate base system. Based on this, I aimed to investigate fat creams prepared using a sugar alcohol and an intense sweetener.

Material and method

In accordance with my objectives, I included the following fats, carbohydrates, and sugar substitutes in the experiments.

Fats**:** Palm fat, coconut fat (lauric fat), milk fat (animal-derived fat)

Carbohydrates**:** Fructose (monosaccharide), sucrose (disaccharide, in two particle sizes), inulin (oligosaccharide), starch (polysaccharide)

Sugar substitutes: Xylitol (sugar alcohol), stevia (intense sweetener).

The fats and carbohydrates were mixed in all cases at a 50% weight ratio at room temperature. The sugar substitutes were incorporated according to their sweetening power: xylitol at a 50% weight ratio, and stevia at a 20% weight ratio (20% stevia – 80% palm fat).

For the characterisation of the raw materials, I performed measurements of fatty acid composition and particle size distribution. For the prepared systems, I examined the solid content using an NMR device at various temperatures, the apparent viscosity using a rotational viscometer at 30°C, and the texture of the samples using a penetrometer for structural analysis. Additionally, I investigated their thermal behaviour using a DSC device.

In each test, multiple parallel measurements were conducted, and the results were evaluated using statistical analysis. This included MANOVA, ANOVA, Tukey post hoc tests, Pearson correlation, and linear discriminant analysis.

Results and discussion

Based on the experimental results, it can be concluded that sugar increases the solid fat content in pure fats. Considering the different particle sizes, it can be said that reducing particle size increases the SFC (Solid Fat Content) value, likely because larger sugar particles provide fewer active sites due to their smoother surface, thereby hindering recrystallisation. For different carbohydrates, the measured SFC values clearly show that lower temperatures result in different SFC values. Correlation analysis suggests that the results correlate more with carbohydrate chain length than with particle size.

Viscosity measurements indicate that the presence of carbohydrates increases the apparent viscosity and shear stress, regardless of fat type. Smaller particle size results in higher viscosity. Shear stress values are proportional to carbohydrate chain length, being highest with starch and lowest with fructose and sucrose. It is not possible to draw a definitive conclusion as to whether carbohydrate chain length or particle size has a greater effect on the viscosity of palm fat. For all samples, the Herschel-Bulkley model provided the most accurate fit among the selected rheological models.

Texture measurements show that the addition of sugar increased the hardness of the samples, and consequently, the work required to compress them, across all fat samples. In terms of adhesion, the addition of sugar increased the values for coconut and milk fat, while it decreased for palm fat. It can be concluded that increasing particle size led to increased hardness. For adhesion, the smaller the particle size, the lower the measured value. For different carbohydrates, each addition caused an increase in hardness and work values compared to pure palm fat. With the growth of the carbon chain in mono-, di-, and oligosaccharides, the values proportionally increased, but the sample made with starch as a polysaccharide did not follow this trend. The results for this sample fell between those of fructose and sucrose. When examining adhesion in these samples, no clear trend could be observed regarding the effect of carbohydrates. Compared to the palm fat value, the measured value was lower for fructose, sugar, and inulin, but higher for starch.

Based on the DSC (Differential Scanning Calorimetry) results, it can generally be said that the presence of sugar reduced the areas under the curve, i.e., the enthalpy values, regardless of fat type. Peaks became harder to detect, and the corresponding temperatures shifted in both crystallisation and melting. During crystallisation, the presence of sugar shifted peak detectability towards higher temperatures in all samples. During melting, higher temperatures were observed in palm fat, while lower temperatures were recorded in the other two samples. The melting of sucrose always appeared as a well-defined peak. Examining the temperature range of crystallisation and melting, it can be concluded that crystallisation begins at a lower temperature, thus occurring later in the presence of sugar, but ends at a higher temperature, hence earlier. Based on these findings, it can be inferred that the presence of sugar inhibits the formation of crystal nuclei but promotes crystal growth. The melting of fat crystals begins at a lower temperature due to the effect of sugar and ends at a lower temperature in milk fat and coconut oil, but at a higher temperature in palm fat. Reducing particle size resulted in a decrease in enthalpy values. Based on the different carbohydrates, it can be concluded that each carbohydrate led to a decrease in enthalpy and a shift in peaks during the crystallisation and melting of palm fat.

For the sugar substitutes, both xylitol and stevia showed lower values for hardness and penetration work compared to the sugar samples, while adhesion increased compared to the sugar sample. For apparent viscosity and shear stress, the following descending order can be established: xylitol, palm, sugar, and stevia. In this case, too, the Herschel-Bulkley model provided the most accurate fit. Based on the DSC measurements, the enthalpy-reducing effect observed with sugar was also seen in the xylitol and stevia samples. Sugar had the greatest effect in reducing enthalpy compared to pure palm fat, followed by xylitol, and lastly stevia. Overall, the results obtained for the sugar substitutes show that the xylitol sample exhibited values most similar to those of sugar.

In conclusion, based on my doctoral work, it can be determined that carbohydrates influence the melting, crystallisation, texture, and viscosity of fats, regardless of the type of fat. Furthermore, it is evident that the type and particle size of the carbohydrate, as well as the sweetener used, have a significant impact on these parameters. The model experiments I conducted contribute to the development of lipid-carbohydrate-based products. They assist in the selection of appropriate manufacturing parameters and in the formulation of the proper composition and recipe to achieve the desired properties of the final product.

Conclusions and proposals

Lipid-carbohydrate-based products constitute a significant proportion of confectionery products. However, most literature related to this topic focuses predominantly on pure lipids and multi-component complex foods. In the case of the latter, the effects of sucrose are typically the only ones investigated. Based on my research, it is evident that examining different carbohydrates individually and breaking down the food matrix into its components is both worthwhile and necessary.

Measurements conducted with sucrose revealed that, regardless of the type of fat, the presence of sucrose resulted in an increase in most measured parameters compared to the values observed for pure fats. It increased solid fat content, apparent viscosity, shear stress, hardness, and the work required for compression. The only decrease was observed in enthalpy values. The addition of sucrose yielded varying results for adhesion parameters depending on the type of fat. For viscosity modelling, the Herschel-Bulkley model proved to be the most suitable for fitting the data.

When investigating sucrose with two different particle sizes, it was found that reducing particle size increased solid content and viscosity, while decreasing hardness, adhesion, and enthalpy. Consequently, the choice of particle size is a critical parameter for both technological processes and the final product.

The addition of carbohydrates with different chain lengths resulted in increased solid content, viscosity, hardness, and penetration/compression work. Enthalpy values decreased compared to pure fat. Adhesion parameters showed both increases and decreases depending on the carbohydrate type. The effects of the applied carbohydrates on fats were not uniform, highlighting their importance in achieving the desired physical properties (e.g., product hardness) and manufacturing parameters (e.g., flow behaviour and pumpability).

Sweeteners also influenced the aforementioned properties. It is important to emphasise that while these are used to replace sucrose in flavour, the experimental results indicate that they do not have identical techno-functional effects to sugar, either in manner or magnitude. Both xylitol and stevia showed lower values for hardness and compression work, whereas adhesion force and melting enthalpy exhibited higher values.

The xylitol mixture produced higher apparent viscosity values compared to sucrose but demonstrated nearly identical results for melting and crystallisation temperatures as well as crystallisation enthalpy. In contrast, the stevia mixture resulted in lower apparent viscosity values. The melting temperatures were similar to those of the sucrose sample, while crystallisation enthalpy values were higher, and crystallisation temperatures were lower.

In conclusion, the utilisation of lipid-carbohydrate systems in complex foods requires further investigation. It would be worthwhile to continue studying the effects of various carbohydrates with identical particle sizes, as well as to examine other mixing ratios.

New scientific results

1. For the mixtures of coconut fat and milk fat with sucrose, as well as the mixtures of palm fat with carbohydrates other than sucrose, I was the first to demonstrate that the addition of carbohydrates to the fat system influences the development of solid fat content (SFC). This, in turn, affects the melting properties of the samples and subsequently impacts production parameters in industrial processes. The addition of different carbohydrates resulted in varying SFC values at lower temperatures. It can be concluded that, although particle size influences SFC values, as observed in the case of sucrose, the primary determining factor here was the carbohydrate chain length.

2. For lipid-carbohydrate systems, I was the first to establish that, based on apparent viscosity measurements at 30°C, the Herschel-Bulkley model provided a more accurate fit than the Casson model, regardless of changes to the components.

3. I was the first to demonstrate that for fine-grained sucrose and palm fat systems, the Herschel-Bulkley model was a better fit. This indicates that reducing particle size improves the accuracy of the fit. For coarse-grained sucrose, the coefficient of determination was 0.91, the estimation error

was 5.26 Pa, and the error percentage was 6.58%. In contrast, for fine-grained sucrose, these values were 1.0, 0.01 Pa, and 0.01%, respectively.

4. For systems containing only sucrose and fat, I was the first to determine that increasing particle size leads to an increase in the hardness parameter but a decrease in spreadability. As the carbon chain length increases, the hardness and compression work values also increase proportionally in the case of mono-, di-, and oligosaccharides. However, the starch-based sample (polysaccharide) did not follow this trend.

5. For coconut fat and milk fat, I was the first to demonstrate that sucrose induces the melting of fat crystals at a lower starting temperature and causes the melting process to finish at a lower temperature as well.

6. I was the first to confirm that carbohydrates other than sucrose and sucrose crystals with varying particle sizes lead to a reduction in enthalpy and a shift in peak temperatures during the crystallisation and melting of palm fat. No correlation was observed between carbohydrate chain length and changes in DSC results.

7. Sugar substitutes, similar to sucrose, resulted in lower enthalpy values compared to pure palm fat. While stevia decreased hardness and viscosity values relative to pure fat, xylitol increased them.

Publications

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