JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY

Effect of Different Dextrose Equivalent of Maltodextrin on the Interactions with Anionic Surfactant in an Isothermal Titration Calorimetry Study

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Isothermal titration calorimetry (ITC) was used to study interactions between an anionic surfactant (sodium dodecyl sulfate, SDS) and maltodextrins with different dextrose equivalents (DE) in a buffer solution (pH 7.0, 10 mM NaCl, 20 mM Trizma, 30.0 °C). The interaction between SDS and maltodextrin was exothermic, which was attributed to incorporation of the hydrocarbon tail of the surfactant into a helical coil formed by the maltodextrin molecules. ITC measurements indicated that the number of SDS molecules bound per gram of maltodextrin increased with decreasing maltodextrin DE, i.e., increasing molecular weight. It was proposed that SDS only binds to maltodextrin molecules that have a DE greater than 10 glucose units.

KEYWORDS: Isothermal titration calorimetry; maltodextrin; surfactant; binding

INTRODUCTION

Interactions between water-soluble biopolymers and surface-active lipids have attracted great interest because they lead to the formation of complexes that are important in many industrial and natural processes. Several water-soluble uncharged polymers interact with ionic surfactants in solution and thereby give the system unique rheological characteristics, e.g., formation of thermoreversible gels, alteration of transition temperatures, or changes in solution viscosity or gel strength (*1*, *2*). This phenomenon offers an interesting feature for pharmaceutical formulations, since many drug molecules have an amphiphilic character and polymers are commonly used in preparations as thickeners and flow modifiers (*3*). In addition, surfactants alter the conformation and self-association of biopolymers in aqueous solutions, which leads to changes in the appearance, stability, and rheology of the solution (*4*−*11*). Changes in the conformation or aggregation of biopolymer molecules may lead to an appreciable change in their functional attributes, e.g., surface activity, thickening, or gelation (*12*).

In this paper, we are primarily interested in interactions between surface-active lipids and carbohydrates. These interactions are usually noncovalent in nature, such as ionic, hydrophobic, or hydrogen bonding. There are generally two types of interactions: the molecular "binding" of individual lipid molecules to the carbohydrate and the interaction between a lipid "phase" (micelles) and the carbohydrate (*13*). However, the main driving force for both of these interactions is the hydrophobic interaction, involving the surfactant chains (*14*). Amylose−lipid complexes are one of the most well understood examples of interactions between polysaccharides and individual polar lipid molecules. Complex formation is driven by the hydrophobic effect, i.e., the amylose helix offers a better environment for the hydrocarbon chain than the aqueous surrounding (*13*). In addition, there is evidence that amylose can interact strongly with many polar and nonpolar compounds, including lipids and emulsifiers (*15*−*19*). At the level of secondary structure, the interaction is believed to involve the incorporation of lipid molecules within single-helical conformations of the amylose (*20*). Moreover, different studies of lipid-binding (equilibrium dialysis and surface tension measurements) showed that polar lipids are also "bound" to amylopectin or waxy starches (*21*, *22*). Equilibrium dialysis studies have shown that normal maize bound approximately 7 times more lipid than waxy maize (*21*). Surface tension measurements gave a surfactant−polysaccharide binding ratio that was 17.5 times higher for amylose than amylopectin (*22*, *23*). In the former case, the binding of stearic acid was studied and in the latter SDS.

In this study, maltodextrin was used as a model system. Maltodextrins contain linear amylose and branched amylopectin degradation products from enzymic hydrolysis of starches (*24*). They represent a mixture of saccharides with a broad molecular weight distribution, depending on dextrose equivalent (DE), which reflects the degree of hydrolysis. Higher DE leads to a decrease in average molecular weight and a change in physicochemical properties. Hygroscopicity, solubility, osmolality, and their effectiveness to reduce the freezing point increase with increasing DE, while viscosity, cohesiveness, and coarse-crystal prevention increases with decreasing DE (*25*). Maltodextrins are more water soluble than starch. Reviews of the applications of maltodextrins in the food industry have been given elsewhere (*24*, *26*−*31*). Linear chains of maltodextrin and starch molecules can form helical complexes upon interacting with hydrophobic

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10.1021/jf034052u CCC: $25.00
Published on Web 11/21/2003

tails of amphiphilic lipid molecules (*32*). These polysaccharide−lipid complexes are believed to have different molecular and physiochemical properties than the polysaccharide alone.

One of the objectives of our research is to understand how polar lipids interact with long-chain glucose polymers (e.g. amylose and amylopectin) and how these interactions change the functional characteristics of starch and its derivatives. An improved understanding of the origin and nature of the interactions between amylose/amylopectin and surface-active lipids may lead to the design of new foods with improved nutritional, physicochemical, and sensory properties.

MATERIALS AND METHODS

Materials. Maltodextrin with DE 5 (MALTRIN 40), DE10 (MALTRIN 100), DE15 (MALTRIN 150), DE20 (MALTRIN 200), and DE25 (MALTRIN 250) were obtained from Grain Processing Corp. (Muscatin, IA). Sodium dodecyl sulfate (SDS) and Trizma base were purchased from Sigma Chemical Co. (St. Louis, MO), sodium chloride was from Mallinckrodt Baker, Inc. (Paris, KY), and hydrochloric acid was from Fisher Scientific (Fair Lawn, NJ). Deionized and distilled water was used for the preparation of all solutions.

Solution Preparation. A stock buffer solution (pH 7.0, 10 mM NaCl, 20 mM Trizma) was prepared by dispersing Trizma base and sodium chloride into water and then adjusting the pH with hydrochloric acid solution. Maltodextrin (0.5% w/v) solutions were prepared by dispersing maltodextrin in stock buffer solution and stirring for 60 min. SDS (35 mM) solutions were prepared by dispersing SDS into stock buffer solution and stirring for 60 min. The pH of SDS−maltodextrin solutions decreased by less than 0.16 units when SDS solution was titrated into maltodextrin solution under similar conditions as those used in the ITC experiments.

Isothermal Titration Calorimetry. An isothermal titration calorimeter (VP-ITC, Microcal Inc., Northampton, MA) was used to measure enthalpies of mixing at 30.0 °C. Aliquots (10 μL) of 35 mM SDS solution were injected sequentially into a 1480-μL reaction cell initially containing either buffer solution or maltodextrin solution. Each injection lasted 20 s and there was an interval of 300 s between successive injections. The solution in the reaction cell was stirred at a speed of 315 rev min^{-1} throughout the experiments. All solutions were degassed prior to the measurements being carried out.

RESULTS AND DISCUSSIONS

SDS Micellization in Buffer Solutions. Initially, ITC was carried out to establish the critical micelle concentration (cmc) of the SDS in the buffer solutions used in this study. A heat flow versus time profile resulting from sequential injections of 10-μL aliquots of surfactant solution (35 mM SDS, pH 7.0, 10 mM NaCl, 20 mM Trizma) is shown in **Figure 1a**. The surfactant concentration in the injector was above the cmc, so the injector contained a mixture of micelles and monomers. Initially, a series of relatively large endothermic peaks was observed when the surfactant solution was injected into the reaction cell. These enthalpy changes are the result of micelle dissociation, because the surfactant concentration in the reaction cell was initially below the cmc (*33*). After a certain number of injections, there was an appreciable decrease in peak height, because the surfactant concentration in the reaction cell exceeded the cmc and so the micelles injected into the reaction cell no longer dissociated. Above the cmc the enthalpy change is therefore only the result of micelle dilution effects (*33*). The dependence of the enthalpy change per mole of surfactant (ΔH/Δ[SDS]) injected into the reaction cell on the surfactant concentration in the reaction cell was calculated by integration of the heat flow versus time profiles (**Figure 2**). The cmc of the SDS was determined from the inflection point in the ΔH/

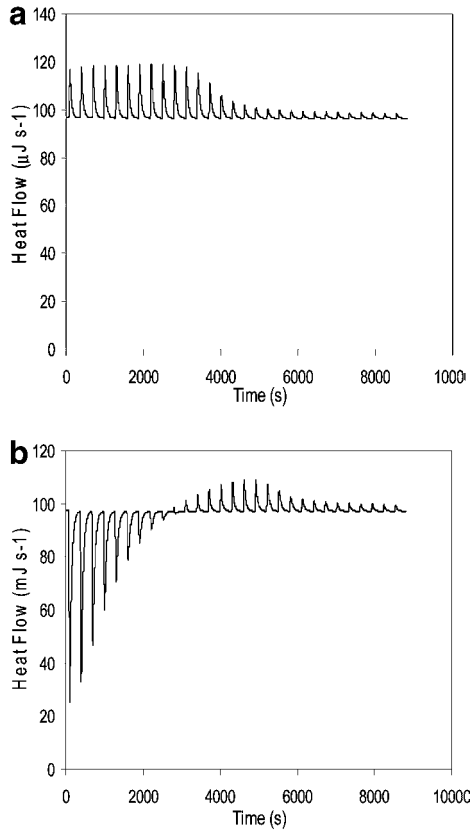

Figure 1. (**a**) Heat flow vs time profiles resulting from injection of 10-μL aliquots of 35 mM SDS into a 1480-μL reaction cell containing buffer solution at 30.0 °C. (**b**) Heat flow vs time profiles resulting from injection of 10-μL aliquots of 35 mM SDS into a 1480-μL reaction cell containing 0.5% (w/v) maltodextrin DE5 solution at 30.0 °C.

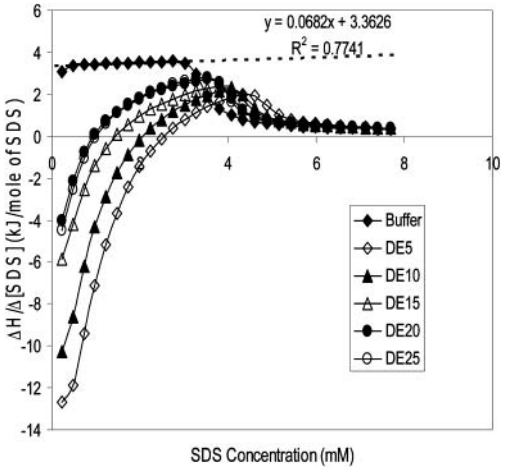

Figure 2. Dependence of enthalpy on SDS concentration in maltodextrin solutions at 30.0 °C.

Δ[SDS] versus surfactant concentration curve as 3.52 ± 0.04 mM, which was close to the cmc (3.4 ± 0.1 mM) of SDS determined by ITC in our previous study (*34*).

Effect of DE on SDS−Maltodextrin Interactions. In the absence of maltodextrin, the enthalpy change resulting from the injection of SDS into buffer solution was large and endothermic below the cmc because of micelle dissociation, but it was relatively small above the cmc because of micelle dilution effects (**Figures 1a** and **2**). In the presence of maltodextrin, there was a large exothermic contribution to the enthalpy change below the cmc (**Figures 1b** and **2**). To highlight the enthalpy changes

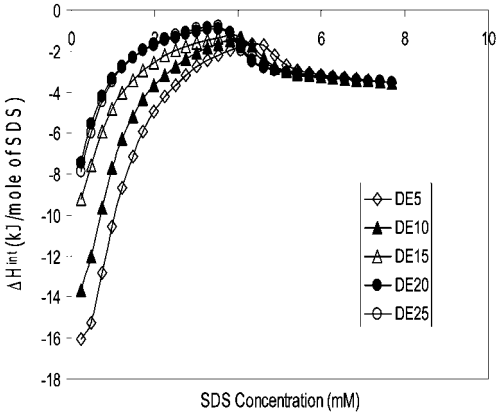

Figure 3. Enthalpy changes due to SDS–maltodextrin interactions (ΔH_{int}) as a function of SDS concentration. SDS was injected into maltodextrin solutions in a reaction cell at 30.0 °C.

due to SDS−maltodextrin interactions (ΔH_{int}), we subtracted the demicellization enthalpy (ΔH_{demic}) from the overall measured enthalpy ($\Delta H_{int} = \Delta H - \Delta H_{demic}$) (**Figure 3**). The demicellization enthalpy of SDS was determined from the mean of the ITC measurement made below the cmc (i.e. SDS = 0.3 → 3 mM) to be $\Delta H_{demic} = 3.4$ kJ mol^{-1}. We propose that the large exothermic peaks were the result of SDS−maltodextrin interactions.

The enthalpy changes associated with these interactions could have been due to binding of SDS to maltodextrin or due to changes in the conformation of the maltodextrin (e.g., a coil-to-helix transition). However, it is not possible to identify the physical origin of the enthalpy changes from ITC measurements alone. The enthalpy change upon injection of SDS into the reaction cell was highly dependent on the dextrose equivalent (DE) of maltodextrin (**Figure 3**). Nevertheless, the enthalpy change versus surfactant concentration profiles followed a fairly similar pattern for all DE of maltodextrin. At low SDS concentrations, the enthalpy change was highly exothermic. As the SDS concentration was increased, the enthalpy change became less exothermic, until it eventually reached a maximum value. After the maximum, the enthalpy change became increasingly exothermic again and eventually tended toward the value measured in the absence of maltodextrin.

The observed changes in the enthalpy profiles can be explained in terms of the interaction of surfactant molecules with the maltodextrin. At relatively low surfactant concentrations, there was a highly exothermic reaction due to binding of SDS molecules to the maltodextrin. As the surfactant concentration increased the number of available binding sites on the maltodextrin decreased; hence, the exothermic contribution to the enthalpy change of SDS−maltodextrin interaction was decreased. Eventually, all of the binding sites on the maltodextrin became saturated and, therefore, any further SDS micelles injected into the reaction cell did not interact with the maltodextrin. Consequently, the enthalpy changes associated with SDS−maltodextrin interaction at high surfactant concentrations remained constant and similar to those determined in the absence of maltodextrin. When the concentration of free SDS monomers in the aqueous phase increased above the cmc, the corrected enthalpy changes were due to micelle formation (since enthalpy changes due to demicellization have been subtracted from the experimentally measured values).

An apparent critical micelle concentration (cmc*) for each of the maltodextrin−SDS solutions was determined from the inflection point in the $\Delta H/\Delta$[SDS] versus [SDS] curves that

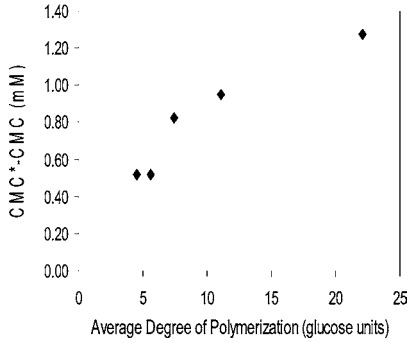

Figure 4. Apparent critical micelle concentration of maltodextrin–SDS solutions as a function of degree of polymerization of maltodextrin at 30.0 °C.

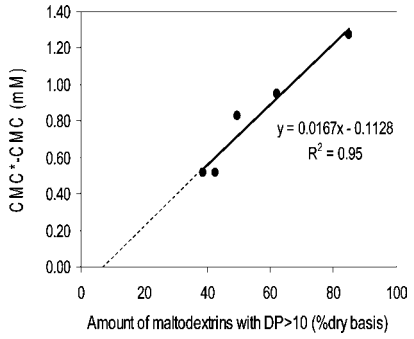

Figure 5. Apparent critical micelle concentration of maltodextrin–SDS solutions as a function of the amount of maltodextrins with DP > 10 (% dry basis) at 30.0 °C.

occurred after the endothermic peak shown in **Figure 2** (**Figures 4** and **5**). The difference between the cmc of the surfactant in the presence and absence of maltodextrin (Δcmc = cmc* − cmc) should be equal to the amount of surfactant that binds to the maltodextrin at saturation (*34*). There was an increase in Δcmc with decreasing DE of maltodextrin, i.e., with increasing degree of polymerization of glucose units (**Figure 4**). The Δcmc values of DE 20 and 25 were fairly similar, because they had similar average degrees of polymerization (**Table 1**).

As mentioned earlier, there are believed to be two general types of surfactant−polysaccharide interaction: (i) binding of individual lipid molecules to polysaccharides, e.g., through inclusion in a helix, and (ii) formation of surfactant micelles on polysaccharide backbones (*13*). The SDS−maltodextrin interactions in our study are most likely to be due to formation of helical inclusion complexes because the interactions occurred below the cmc. The binding of amphiphilic lipids to glucose polymers results in the formation of a complex in which the hydrophobic tail of the lipid is surrounded by a helix of glucose monomers (*20*, *32*). The hydrocarbon tail of a SDS molecule is approximately 1.7 nm long (*35*). Previous studies indicate that linear alcohols and fatty acids form helices with six D-glucosyl residues per turn (*36*), which have an average length of about 0.8 nm per turn (*37*). The alkyl chains of SDS molecules have been shown to fit into helices of about 12−16 glucose units, corresponding to a length of about 1.6−2.0 nm. It is therefore possible that only those maltodextrin molecules in the solution that have a degree of polymerization around 12 or higher may have sufficient glucose units to completely surround the hydrocarbon tail of SDS and form stable helices.

Since maltodextrin ingredients generally contain a range of glucose polymers with different molecular weights (**Table 1**), the plot between Δcmc and the average degree of polymerization of glucose units (**Figure 4**) does not give precise binding

Table 1. Compositions[a] of Maltodextrin Ingredients with Different DE (Dextrose Equivalent) Values Used in This Study

degree of polymerization[c]	composition[b] (%)				
	DE 5	DE 10	DE 15	DE 20	DE 25
1 (glucose)	0.3	0.8	1.3	2.3	7.6
2 (maltose)	0.9	2.9	4.8	7.4	6.9
3	1.4	4.4	6.7	9.1	7.0
4	1.4	3.8	5.5	6.8	6.8
5	1.3	3.4	4.7	6.3	6.3
6	1.8	5.7	8.4	11.9	5.6
7	2.4	6.8	9.1	10.0	5.1
8	2.0	4.5	4.8	3.7	4.6
9	1.8	3.1	2.9	2.1	4.1
10	1.7	2.5	2.1	1.7	3.4
+10	85.0	62.1	49.7	38.7	42.6
av theoretical MW[d]	3600	1800	1200	900	720
av DP[c]	22.1	11.1	7.4	5.6	4.5

[a] Compositions are reported as the amount (% dry basis) of maltodextrin in classes with different degrees of polymerization (DP), as determined by high-performance liquid chromatography (HPLC) (modified from bulletin of Grain Processing Co., Muscatin, IA). [b] Dextrose equivalent (DE) is a measure of the total reducing power of all sugars present relative to glucose as 100 and expressed on a dry weight basis. [c] Degree of polymerization (DP) of glucose units. [d] Molecular weight.

parameters. To support our hypothesis that SDS would only bind to maltodextrin molecules above a certain molecular weight, we plotted Δcmc versus the amount of maltodextrin with DE >10 (**Figure 5**). We found that this plot was linear ($r^2 = 0.95$) and passed close to the origin, which supports the notion that SDS only binds to longer maltodextrin molecules (*32*, *34*, *38*−*40*).

CONCLUSIONS

This study has shown that isothermal titration calorimetry measurements can provide useful information concerning interactions between maltodextrin and anionic surfactants. According to this method, the binding of SDS to maltodextrin increased with decreasing DE (degree of polymerization). The interaction only seemed to occur with maltodextrin molecules greater than about 10 glucose units. It was proposed that the interaction was due to the insertion of the nonpolar tail of the surfactant into a helical coil formed by the maltodextrin molecules. Nevertheless, further work using other analytical techniques is required to conclusively establish the molecular basis of this interaction. Work in our laboratory is currently underway to investigate interactions between maltodextrin and other types of surfactant.

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Received for review January 17, 2003. Revised manuscript received October 14, 2003. Accepted October 28, 2003. This material is based upon work supported by the Cooperative State Research, Extension, Education Service, United States Department of Agriculture, Massachusetts Agricultural Experiment Station (Project No. 831).

JF034052U