

Phase Behavior and Component Partitioning in Low Water Content Amorphous Carbohydrates and Their Potential Impact on Encapsulation of Flavors

Yvonne M. Gunning,[†] Roger Parker,^{*,†} Steve G. Ring,[†] Neil M. Rigby,[†] Benjamin Wegg,[†] and Anthony Blake[‡]

Food Materials Science Division, Institute of Food Research, Norwich Research Park, Colney, Norwich NR4 7UA, United Kingdom, and Corporate R&D Division, Firmenich S.A., 1 Routes des Jeunes, CH-1211 Geneva 8, Switzerland

The compositions at which amorphous ethanol–maltose–water mixtures exhibit liquid–liquid separation have been determined in the temperature range from 20 to 80 °C. At water contents below ~20% w/w two phases were observed, with the maltose-rich phase slightly richer in water. Partition coefficients of organic nonelectrolytes ranging in hydrophobicity from 1,2-ethanediol and 1,2-propanediol to benzyl alcohol and propyl acetate have been measured for octanol/sorbitol, benzyl alcohol/sorbitol, and 1-butanol/sorbitol mixtures. Linear correlations were found between the log partition coefficients in the various solvent systems. Replacing water with sorbitol results in more organic partitioning into the octanol. Replacing octanol with benzyl alcohol or 1-butanol also results in more organic partitioning into the hydrophobic phase. The results establish a relationship with partition coefficients for octanol/water mixtures, which are well studied experimentally and for which predictive approaches exist. The implications of these results for flavor retention and encapsulation are discussed.

Keywords: *Flavor; partitioning; carbohydrates; log P*

INTRODUCTION

Liquid flavor systems are encapsulated in low water content glass-forming carbohydrates to prevent their evaporation, to protect them from adverse chemical reactions, and to convert them into easily handled free-flowing powders (Whorton, 1995). Typically, glassy carbohydrate mixtures, such as sucrose and maltodextrin with water contents <5% w/w, are used as encapsulation matrices. To function, the carbohydrate encapsulation matrices must, to a large extent, be impermeable to the components of the flavor system. Furthermore, the flavor must not be so soluble in the matrix that the particles become sticky and liable to caking, a behavior associated with surface tension driven viscous flow of the matrix when it is above its glass transition (Wallack and King, 1988; Slade and Levine, 1995). Thus, the phase behavior and component partitioning in flavor–carbohydrate–water mixtures is a key aspect in the design of encapsulation systems.

Few experimental studies have been made to determine the solvent qualities of low water content amorphous carbohydrates. Dry amorphous carbohydrates are hygroscopic and will readily absorb water from the atmosphere, a behavior that is quantified in terms of water sorption isotherms (Roos, 1993a; Moates et al., 1997). The amount of water absorbed increases steadily with increasing external water activity, indicating that water and amorphous carbohydrates are completely

miscible. Partial miscibility of water with the carbohydrate would be indicated by a region of increasing absorption at constant water activity because at thermodynamic equilibrium the water in the two coexistent phases must have the same water activity [see, for example, Lam and Benoit (1974)]. Recently, the temperature dependence of the sorption isotherms was used to characterize the solvent quality of carbohydrate–water systems via the solubility parameter approach developed by Hildebrand, Scott, and Scatchard (Benczédi et al., 1998a,b). Only isolated pieces of information are available on the miscibility of amorphous carbohydrates with the components of flavor systems. Simplistically, considering their polar nature and hydrogen-bonding capabilities, it might be expected that liquid carbohydrates would have solvent qualities similar to those of water. However, in a study of the ternary phase diagram of acetone–water–sucrose mixtures Verhaar reported (Bubnik and Kadlec, 1995) that, for certain compositions, at water contents below ~37% w/w, phase separation of acetone-rich and sucrose-rich phases occurs. Thus, although acetone and water are fully miscible, acetone and sucrose are only partially miscible, and, so, it can be concluded that carbohydrate liquids are not as good solvents for moderately hydrophobic species as water. In the present study, the miscibility of ethanol and maltose in the presence of water is studied as a model for the behavior of a flavor–carbohydrate–water mixture. In these experiments the flavor is being modeled by a single component, ethanol, and the effect of water content on the compatibility with a carbohydrate matrix former is characterized. Ethanol–maltose–water mixtures were used as models in earlier work on the retention of volatile species in the

* Author to whom correspondence should be addressed (telephone 44-1603-255396; fax 44-1603-507723; e-mail roger.parker@bbsrc.ac.uk).

[†] Institute of Food Research.

[‡] Firmenich S.A.

freeze-drying of carbohydrates (Flink and Karel, 1970). Ethanol models a small polar molecule that could, potentially, permeate a low water content carbohydrate encapsulation matrix. A nonequilibrium ternary phase diagram for ethanol–maltose–water is reported describing solely the amorphous phases at low water contents. The occurrence of metastable amorphous maltose-rich phases is recorded by studying the mixtures over time scales shorter than required for the nucleation of a crystalline maltose phase.

Another aspect of this problem is that liquid flavor systems are multicomponent mixtures composed of diverse species ranging from nonpolar hydrophobic molecules to polar and ionic hydrophilic species. When a flavor is mixed with water, the low solubility of the more hydrophobic components means that they form a separate phase, often in a dispersed state, as an emulsion. Depending upon the relative affinities for the two phases, the components of the flavor system partition between them. In medical, pharmaceutical, and agrochemical research, partitioning between hydrophobic and hydrophilic phases has been extensively studied (Leo et al., 1971; Leo, 1991; Sangster, 1997). Partitioning in the octanol–water system is used as a benchmark. Considerable progress has been made in predicting partition coefficients from molecular structure (Leo, 1993), and commercial software packages are available to perform these predictions. To relate the partitioning of components in carbohydrate-based flavor encapsulation systems to the behavior in octanol–water mixtures, a series of partition coefficients were measured using a liquid carbohydrate as the hydrophilic solvent and octanol, benzyl alcohol, or 1-butanol as the hydrophobic solvent. A series of organic compounds selected to be easy to handle and assay and to exhibit a range of partition coefficients is used to model the components of the flavor system. In these experiments it is the combination of the hydrophobic solvent and the organic component being partitioned that models the liquid flavor system.

MATERIALS AND METHODS

Materials. Ethanol (Analar), formamide (Analar), Karl-Fischer reagent (pyridine free), 1-propanol, ethyl acetate, octanol, and methanol (Analar) were all obtained from BDH Chemicals (Lutterworth, Leicestershire, U.K.). Maltose monohydrate (grade 1) was obtained from Sigma Chemicals (Poole, Dorset, U.K.). Sodium tartrate dihydrate (Karl-Fischer standard) was obtained from Fluka Chemicals (Gillingham, Dorset, U.K.). 1,2-Ethanediol, 1,2-propanediol, 2,3-butanediol, diethyl tartrate, triacetin, triethyl citrate, benzyl alcohol, propyl acetate, and sorbitol were obtained from Aldrich (Gillingham, Dorset, U.K.). The water for HPLC was from an Elga "Ultra Pure" water purifier. All other water was distilled deionized water.

Preparation of Amorphous Ethanol–Maltose–Water Mixtures. Concentrated aqueous solutions of the maltose (5–20% w/w water) were prepared by heating 2 g samples to 130 °C in screw-topped Pyrex culture tubes in a preheated hot-block, with occasional mixing with a vortex mixer. These were cooled prior to the addition of the ethanol and were then gently reheated and remixed.

Determination of Phase Diagram. Ethanol–maltose–water mixtures were left to equilibrate for at least 3 days at 20, 40, 60, and 80 °C. After preliminary experiments to ascertain the approximate position of the phase boundary, a series of samples were prepared, with the composition systematically varied, each component being a multiple of 5% w/w, and visual inspection was used to establish the occurrence of phase separation. For some mixtures, which readily separated

into two distinct layers, the composition of the coexisting phases was determined using HPLC and Karl-Fischer titration. An HPLC system comprising an Aminex ion exclusion column (300 × 7.8 mm), a Spectra Physics (model 8700) pump, a Gilson refractive index detector (model 131), and a Spectra Physics (model 4270) integrator was used to determine the ethanol and maltose with ultrapure water as the eluent (0.7 mL min⁻¹). The water content was determined by titration using a Karl-Fischer titrator (Baird & Tatlock, model AF5) with 1:1 methanol/formamide as the solvent.

Partition Coefficients. By convention the liquid–liquid partition coefficient, *P*, is defined as the concentration of the species in the hydrophobic phase divided by the concentration of the species in the hydrophilic phase. Samples for determining octanol/sorbitol partition coefficients were prepared by first melting the sorbitol (5 g) by heating to 120 °C in a screw-topped culture tube and then cooling it to 60 °C; the octanol (0.25 mL) and solute to be partitioned (0.1 mL) were then added, and the tube was quickly sealed. The tube was heated to 100 °C, and the two phases were mixed on a Whirlimixer for 2–3 min. The tube was cooled to 70 °C, and the two phases were allowed to separate for 4–5 h. After this time, the mixture was cooled to room temperature and the composition of the organic phase analyzed by GC. This procedure was assumed to be an adequate time for equilibration because a further 24 h for separation at 70 °C and a second mixing at 100 °C did not affect the measured partition coefficient. The partitioning experiment was repeated five times. The solute concentration in the organic phase was determined using a Carlo Erba GC 600 Vega series gas chromatograph with a BPX5 0.32 mm i.d. column (0.5 μm film thickness). The injector and detector temperatures were 260 °C, and the temperature program of the GC oven was as follows: 40 °C for 1 min, heat from 40 to 200 °C at 10 °C min⁻¹, followed by 10 min at 200 °C. The determination of octanol/water partition coefficients was carried out using the above procedure but with no heating of the samples (partitioned at 20 °C). These were compared with the available literature values (Leo et al., 1971). A similar procedure was used for determining partition coefficients with benzyl alcohol and 1-butanol substituted for the octanol. The amount of benzyl alcohol or 1-butanol was increased to 2 mL, and the solute to be partitioned was increased to 0.2 mL. Further experiments using different carbohydrates required some modification of the procedure. Mixtures containing glycerol did not require heating, and those containing maltose, sucrose, and glucose required heating to the appropriate melting temperature (Roos, 1993b).

RESULTS AND DISCUSSION

Phase Behavior. Figure 1 shows the ethanol–maltose–water phase diagram at temperatures from 20 to 80 °C. In the low water content region, phase separation into maltose-rich and ethanol-rich phases is observed. The composition range in which phase separation was observed increases as the temperature is dropped from 80 to 20 °C. The phase boundary is symmetrical about a line that bisects the diagram and passes through the water vertex. Figure 2 shows some tie-lines for mixtures at 40 °C which show that the maltose-rich phase contains more water than the ethanol-rich phase. The viscosity of the maltose-rich phase prevented the coalescence of the ethanol-rich phase at low water contents, not allowing them to be separated for analysis and determination of the tie-lines at lower water contents. The two-phase region is less extensive than in the case of acetone–water–sucrose, occurring only at water contents lower than 25% w/w at 20 °C and 20% w/w at 80 °C, whereas in the acetone–sucrose–water system, separation occurred at water contents less than 40% w/w (Bubnik and Kadlec, 1995).

Although the phase behavior of ethanol–maltose–water exhibits some of the features of a practical

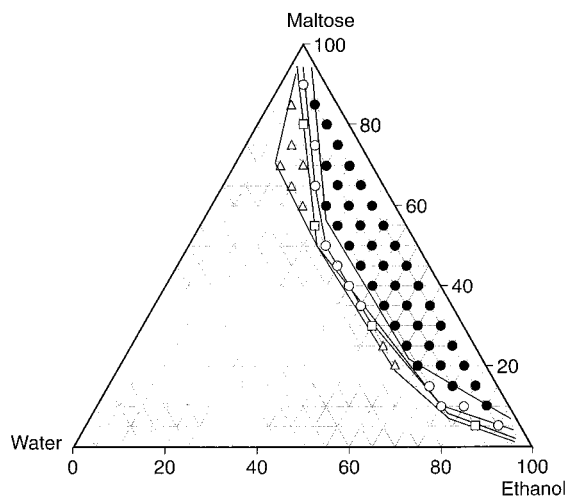


Figure 1. Ternary phase diagram for ethanol–maltose–water: ●, 80 °C; ○, 60 °C; □, 40 °C; △, 20 °C. Crystalline maltose phases omitted and metastable supersaturated maltose phases included. Symbols show maximum temperature at which two liquid phases are observed.

encapsulation system, it also differs in several important ways. In general, the components of flavors encapsulated in amorphous carbohydrates are more hydrophobic than either ethanol or acetone, and so the two-phase region is very much more extensive. The most hydrophobic components that occur in liquid flavor systems are almost wholly immiscible with water. From the above phase diagram it can be predicted that the solubility of the hydrophobic species would be even lower in a low water content amorphous carbohydrate than in water. Furthermore, less water will partition into the hydrophobic phase in more hydrophobic flavors systems, and the water will partition extensively into the amorphous carbohydrate-rich phase. Finally, the water contents of the mixtures in the phase diagram are higher than practical encapsulation systems (3–4% w/w water); consequently, the maltose-rich phase is in a metastable liquid state rather than a glassy state (Noel et al., 1996). In this region of the phase diagram the phase separation would have been arrested by the glass transition of the maltose-rich phase.

The recognition that water can control phase compatibility in flavor–carbohydrate–water mixtures allows some reappraisal of earlier experimental studies in the literature. For example, in their studies of volatile retention in freeze-dried products, the “microregions” described by Flink and Karel (1970) could, in the context of phase behavior, be interpreted as a separate volatile-rich phase that would be dispersed in a carbohydrate-rich phase. Retention could be due to the volatile-rich phase being encapsulated in a carbohydrate-rich phase in which the volatiles are present at relatively low levels. The permeability of the carbohydrate-rich phase to the volatiles depends both on their solubility in the phase and on their diffusive mobility. It is clear from this work that water content has a strong influence on solubility and thereby on permeability. Thijssen’s work on “selective diffusion” (Thijssen, 1971) includes studies comparing the diffusive mobility of acetone and water in low water content maltodextrins. At the lowest water contents studied phase separation of acetone-rich phases may be a factor limiting the apparent diffusivity.

Component Partitioning. The distribution of flavor components between phases is described by a partition coefficient. These have been extensively studied for

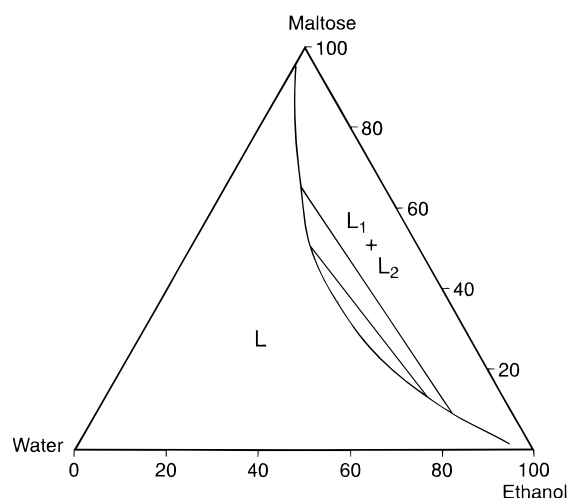


Figure 2. Ethanol–maltose–water phase diagram at 40 °C with tie lines. Regions of phase diagram: L, single liquid phase; $L_1 + L_2$, two coexisting liquid phases.

octanol/water systems and, so, to be able to apply this information to flavor–carbohydrate systems, the initial step was to measure partitioning in an octanol/liquid sorbitol mixture. The octanol models the behavior of the bulk of the flavor, the liquid sorbitol models the behavior of a low water content amorphous carbohydrate, and the species being partitioned represents a minor component of the flavor. Sorbitol was chosen because it can readily be melted to form a liquid that is sufficiently stable with respect to crystallization and chemical reaction to conduct partitioning experiments. Also, the viscosity of the liquid is sufficiently low to allow mixing. The liquids formed from higher molecular weight carbohydrates, for example, sucrose and maltodextrin, which are used in practical encapsulation systems (Gunning et al., 1999), are considerably more viscous than sorbitol, a behavior that can be related to their higher glass transition temperatures, for example, sorbitol, 7 °C, and sucrose, 65 °C (Orford et al., 1990). The species partitioned ranged from the hydrophilic, such as 1,2-ethanediol and 1,2-propanediol, to the moderately hydrophobic, such as benzyl alcohol and propyl acetate. Figure 3 shows a double-logarithmic plot of the partition coefficients in octanol/sorbitol versus those measured in octanol/water. The log partition coefficients, for example, $\log []_{\text{octanol}}/[]_{\text{water}}$, are written in the form $\log P_{\text{octanol/water}}$. Data are given in Table 1. The log partition coefficients for the different solvent pairs show a degree of linear correlation (R^2 , the coefficient of determination = 0.766). The octanol/sorbitol partition coefficients are greater than the corresponding octanol/water coefficients. The difference was greatest for the more hydrophilic species, for example, for 1,2-propanediol, $\log P_{\text{octanol/water}} = -1.41$, $\log P_{\text{octanol/sorbitol}} = -0.09$, which means that P increases by a factor of ~ 20 when water is substituted by sorbitol. This difference becomes smaller for the more hydrophobic species. Overall, replacing water with sorbitol results in higher concentrations of the species being partitioned in the octanol. In other words, sorbitol is a less good solvent for the organic species than water. Although sorbitol is a polyhydroxy compound with extensive hydrogen-bonding capability, it does not have the same solvent quality as water.

The effect of substituting different carbohydrates for the sorbitol on the partitioning of 2,3-butanediol at 70

Table 1. Comparison of Partition Coefficients of Organics between Nonpolar Phases [Octanol, Benzyl Alcohol (BzOH), and 1-Butanol] and Polar Phases (Water and Sorbitol)

organic	log <i>P</i> values octanol/water (at 20 °C)		log <i>P</i> (at 70 °C)		
	lit.	exptl	octanol/sorbitol	BzOH/sorbitol	1-butanol/sorbitol
1,2-ethanediol	-1.93		-1.11 (0.06) ^a	-0.31 (0.02)	-0.29 (0.05)
1,2-propanediol	-1.41		-0.09 (0.28)	0.14 (0.02)	0.20 (0.02)
2,3-butanediol	-0.92		0.34 (0.13)	0.63 (0.03)	0.64 (0.03)
ethanol	-0.32		0.85 (0.09)	1.13 (0.07)	1.19 (0.14)
diethyl tartrate	-0.29		1.24 (0.01)	1.61 (0.15)	1.69 (0.16)
triacetin	0.25		0.57 (0.64)		
1-propanol	0.34		1.29 (0.28)		
ethyl acetate	0.73		1.74 (0.10)		
1-butanol	0.88	0.81 (0.03)	1.48 (0.01)	1.73 (0.14)	
triethyl citrate		0.90 (0.01)	2.18 (0.02)		
benzyl alcohol	1.10	1.14 (0.01)	1.31 (0.01)		1.54 (0.15)
propyl acetate	1.60		1.41 (0.20)		

^a Standard deviations in parentheses.

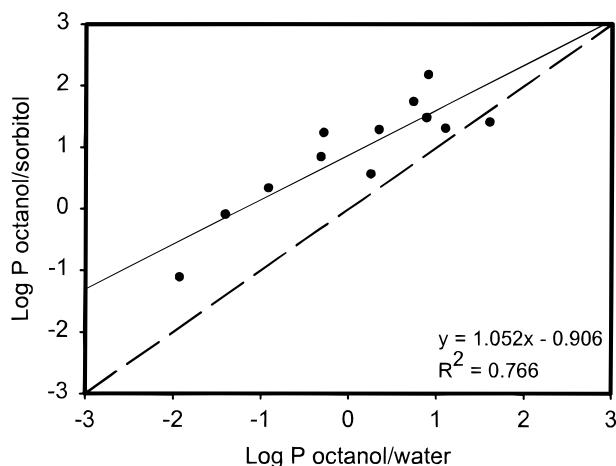


Figure 3. Double-logarithmic plot of octanol/sorbitol partition coefficients versus octanol/water partition coefficient (see data in Table 1). Dashed line shows behavior if partition coefficients were equal.

°C was examined. The log *P* values were as follows: octanol/glycerol, -0.26; octanol/glucose, 0.26; octanol/maltose monohydrate (5% water), 0.28; octanol/sorbitol, 0.31; octanol/sucrose, 0.37. These can be compared with the log *P* value for octanol/water of -0.92. Thus, whereas partitioning in octanol/glycerol has an intermediate value between water and sorbitol, the higher molecular weight saccharides have behavior very similar to that of sorbitol.

The major components of different liquid flavor systems vary in their hydrophobicities. For example, using log *P* octanol/water values to order typical flavor components in terms of their increasing hydrophobicity yields benzyl alcohol (1.10), benzaldehyde (1.47), menthone (2.83*), menthol (3.23*), and limonene (4.35*). The value in parentheses is the log *P* value, either an experimental value or, for those denoted with an asterisk, predicted values obtained using the program ClogP (BioByte Corp., Claremont, CA). For comparison the log *P* of octanol is ~3.0 (experimental value from ClogP database), and so it can be seen that a benzaldehyde-benzyl alcohol mixture, a typical base for a cherry flavor, is less hydrophobic than octanol, whereas a menthone-menthol mixture, a typical base for a peppermint flavor, has a hydrophobicity similar to that of octanol, and a limonene base, typical of a citrus flavor, is more hydrophobic than octanol. The effect of varying the hydrophobicity of the species upon which the liquid

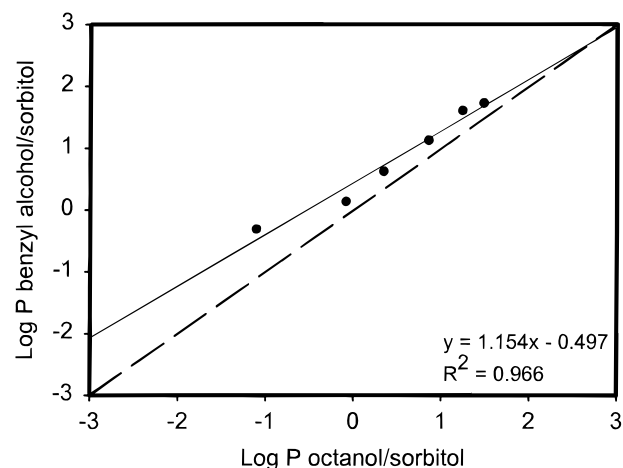


Figure 4. Double-logarithmic plot of benzyl alcohol/sorbitol partition coefficients versus octanol/sorbitol partition coefficient (see data in Table 1). Dashed line shows behavior if partition coefficients were equal.

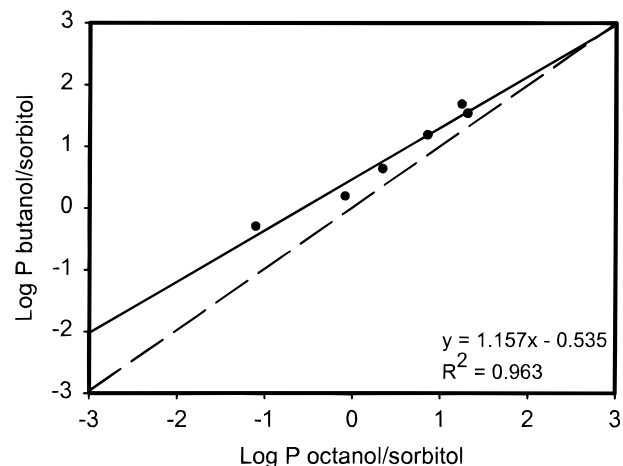


Figure 5. Double-logarithmic plot of 1-butanol/sorbitol partition coefficients versus octanol/sorbitol partition coefficient (see data in Table 1). Dashed line shows behavior if partition coefficients were equal.

flavor systems are based was examined by measuring the partition coefficients of probe species in benzyl alcohol/sorbitol and 1-butanol/sorbitol mixtures. These partition coefficients (Table 1) are plotted against those for octanol/sorbitol in Figures 4 and 5. For the species studied, the log partition coefficients are linearly correlated with R^2 values of 0.966 and 0.963, respectively.

Table 2. Experimental and Predicted Octanol/Water Partition Coefficients at 20 °C for Selected Aroma Compounds^a

compound	exptl log <i>P</i>	calcd log <i>P</i>
diacetyl	-1.34	-1.37
acetaldehyde		-0.224
acetylpyrazine	0.20	-0.497
methional		0.415
methanethiol		0.646
maltol		1.166
4-methoxy-2,5-dimethylfuranone		1.272
vanillin	1.21	1.275
dimethyl sulfide	1.77	1.742
ethyl butyrate		1.77

^a Values from experimental database and predictive calculations of ClogP (Biobyte Corp., Claremont, CA).

In each case there are higher amounts of the probe species being partitioned into the benzyl alcohol and 1-butanol phases than into the octanol phase, which we interpret as being the result of their lower hydrophobicities. A consequence of this partitioning effect is that the hydrophobicity of the major components of a liquid flavor system affect the retention of the more minor components when it is allowed to partition. For example, to maximize the retention of the more polar components of the flavor in the hydrophobic phase, the flavor base should have the lowest hydrophobicity consistent with its remaining incompatible with the carbohydrate phase. For example, basing a liquid flavor system on benzyl alcohol would retain more polar species than one based upon the highly hydrophobic limonene. However, attempts at basing a liquid flavor system upon ethanol, which is more compatible with the carbohydrate matrix than benzyl alcohol, could result in an encapsulation system that would leak and fail to form a glass.

Although the present study has concentrated on the applications of partitioning to flavor encapsulation systems, the concepts could be applied more widely within food and flavor science. The partition coefficients of a range of aroma compounds are shown in Table 2. They are the more polar representatives of the major classes of aroma compounds (Belitz and Grosch, 1999). These compounds have partition coefficients in a range similar to that of the probe species in Table 1. Real foods are almost invariably multiphase, and the phases vary in their hydrophobicity. Using octanol/water partition coefficients as a starting point and then, through judicious experimentation, building a model of how changing the solvent properties of the matrix modifies partitioning is an approach that builds upon the large amount already known about partitioning in octanol/water mixtures. An understanding of how components partition may be particularly useful in the reformulation of foods, for example, the creation of low-fat foods (Schirle-Keller, 1994).

Conclusions. Experiments on the phase and partitioning behavior of model systems indicate that low water content amorphous carbohydrates are less good solvents for flavor components than water. In the ternary system ethanol–maltose–water, the water content controls both whether the system is single phase or two phase and also the extent of the compatibility of ethanol and maltose in the two-phase region. Phase separation and component partitioning may be common in drying processes and should be taken into account. Whenever phase separation occurs, two-phase transport needs to be considered. The partitioning of flavor species depends upon the balance between the interactions in

both the hydrophilic and the hydrophobic phases. In a liquid flavor system the hydrophobicity of the major components, which act as the solvent for the system, influences the partitioning of the minor components of the flavor.

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