SHORT COMMUNICATIONS

Water/Food Flavor Microemulsion Systems

Keywords: Microemulsion; flavors; sodium dodecyl sulfate

INTRODUCTION

Microemulsion is a transparent, isotropic, and thermodynamically stable dispersion, prepared by direct titration to an emulsion system with a medium-chain alcohol. After its introduction by Hoar and Schulman (1943), microemulsion has received much attention and has been reviewed several times (Friberg, 1976; Rosano and Clausse, 1988). These previous papers, however, have been directed toward hydrocarbons as the oilbearing components. Recent interest has focused on triglycerides (Parris et al., 1994) and perfumes (Tokuoka et al., 1994) as the oil component. The latter, such as esters, are an important group of compounds in cosmetic and food processing. Therefore, we believe the investigation of the solubility region of microemulsion with esters, often used as flavors in many food preparations, should fulfill an important need, especially for the food industry.

METHODS AND MATERIALS

Materials. Sodium dodecyl sulfate (SDS) (>99.5%, Mallinckrodt), hexan-1-ol (99%, Merck), ethyl *n*-butyrate (99%, BDH), and ethyl benzoate (99%, BDH) were used as received. Doubly distilled water was used.

Method. A method similar to that employed by Friberg and Gan (1983) was adopted here. The liquid isotropic phases were determined by titration of water of mixtures of the SDS, hexan-1-ol, and flavors in a 7 mm glass tube. The samples were then thoroughly mixed to homogeneity with a vortex meter (Thermolyne Maxi Mix II) and centrifuged (Rexmed Model DSC-1512SDT) at 5000 rpm. The borders were obtained by continuously vortexing selected compositions near phase boundaries and equilibrated for more than a week in a



Figure 1. Solubility region of L_2 in the water/sodium dodecyl sulfate (SDS)/hexan-1-ol system. The line drawn, across the L_2 phase range, toward the water corner indicates the occurrence of maximum water solubility at a weight ratio of SDS to hexan-1-ol of 25/75.

water bath kept at 30 °C. The appearance of the sample was then observed with the naked eye and between cross polarizers. An estimated region of the phases can then be made by observing the clear/turbid transitions.

RESULTS AND DISCUSSION

The alcoholic continuous isotropic region, L_2 , was formed from the combination of water, sodium dodecyl sulfate (SDS), and hexan-1-ol. It was observed to extend from the alcohol apex to about 43 wt % (Figure 1). The maximum water solubility was observed at a weight



Figure 2. Partial pseudoternary phase diagrams for the water, sodium dodecyl sulfate, and a third component consisting of a fixed weight ratio of ethyl *n*-butyrate and hexan-1-ol of $(- \cdot -) 0:1, (-) 1:2, (\cdots) 1:1, and (- -), 2:1.$



Figure 3. Equivalent of Figure 1 but with ethyl benzoate.

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Figure 4. Comparison between the water/food flavor microemulsion regions at weight ratio of flavor to hexan-1-ol of (a) 1:2, (b) 1:1, and (c) 2:1. The solid line represents the ethyl *n*-butyrate system, and the broken line represents ethyl benzoate.

ratio of SDS to hexan-1-ol of 25/75. This value is consistent with the previously reported value (Clausse et al., 1988).

The water/flavor microemulsion regions were determined for compositions with variable flavor/hexan-1-ol weight ratios. The food flavors used are ethyl nbutyrate and ethyl benzoate. Figure 2 shows the solubility region in the water/SDS/hexan-1-ol system with ethyl n-butyrate. The maximum water uptake was maintained with SDS and a flavor/hexan-ol ratio of 25/ 75 and was found to decrease systematically with decreasing hexan-1-ol content. This is an expected result since lesser quantity of hexan-1-ol resulted in fewer associations with the surfactant molecules to form inverse micelle. The maximum water solubility was, however, absent when the composition of the flavor/ hexan-1-ol was greater than 50% (Figure 2). The minimum water solubility, on the other hand, was not affected by the changes.

Figure 3 shows equivalent information but with ethyl benzoate. The association showed a less elongated region and a slight increase in the surfactant ratio for the maximum water uptake. Once again, the region was observed to be decreasing with decreasing alcohol content, and the disappearance of the maximum amount ofwater when the ratio of the flavor/alcohol was greater than 50%. A reduction in the minimum water solubility was also observed as the flavor/alcohol ratio was increased.

The difference in the influence of ethyl *n*-butyrate and ethyl benzoate on the variation of the regions can be seen by a comparison of the respective association regions as shown in Figure 4. Overall, the water/flavor microemulsion region with ethyl *n*-butyrate was observed to be bigger than the corresponding ethyl benzoate region. Similar variations were reported from the pioneering contribution on microemulsion systems with variable hydrocarbon chain lengths of esters (Friberg and Gan, 1983). The explanation for the variations put forth by these authors was due to the equilibrium with lamellar liquid crystal phase. In this investigation, the difference between the flavors is the hydrophobicity of the hydrocarbon chain of their carboxyl group. Hence, the aromatic ring of the carboxyl chain of ethyl benzoate should be expected to exhibit higher hydrophobicity and to stabilize the liquid crystal phase. This results in the reduction of the water/flavor microemulsion region.

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