

Color Changes in Hydrocarbon Oil-in-Water Emulsions Caused by Ostwald Ripening

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The influence of Ostwald ripening on the optical properties of hydrocarbon oil-in-water emulsions stabilized by sodium dodecyl sulfate was investigated. The droplet size, spectral reflectance, and tristimulus color coordinates (L , a , and b) of a series of n -hexadecane and n -octadecane oil-in-water emulsions were measured in the presence and absence of a water-soluble red dye (FD&C Red No. 40). The droplets grew more rapidly in the emulsion containing n -hexadecane than in the emulsion containing n -octadecane because of the higher solubility of n -hexadecane molecules in the aqueous phase. Ostwald ripening caused appreciable changes in n -hexadecane emulsion spectral reflectance and color L , a , and b values due to the growth of emulsion droplets. L , a , and b color values and spectral reflectances of n -octadecane emulsions did not significantly change during the course of the experiment. The results were explained in terms of Ostwald ripening theory and a previously described light scattering theory. The model enables emulsion manufacturers to predict color changes in oil-in-water emulsions that exhibit transcondensational ripening.

Keywords: Emulsion; color; spectral reflectance; light scattering; Ostwald ripening

INTRODUCTION

The perceived quality of many emulsion-based products depends on their color, for example, foods, drinks, paints, pharmaceuticals, and cosmetics. It is therefore of considerable technological importance to understand and to be able to control the factors that influence the optical properties of emulsions. The overall appearance of an emulsion is determined by the way it interacts with light, that is, reflection, transmission, absorption, and scattering (1–4). These interactions are determined by the characteristics of the electromagnetic radiation (wavelength, intensity, and incident angle), the emulsion droplets (size, concentration, and refractive index), and the colorants present (absorption spectra and concentration). Chantrapornchai and coauthors (5, 6) examined the influence of droplet characteristics such as size and concentration and dye characteristics (dye concentration and spectral absorbance) on L , a , and b color values and spectral reflectances of emulsions. Weiss and Liao (7) further examined the influence of the optical properties of the continuous phase on the color and appearance of oil-in-water emulsions. The investigators found that all of these characteristics have a pronounced influence on the light scattering and adsorption behavior of emulsions and thus their color. Processes that are capable of altering any one of these important parameters can therefore be expected to influence the overall appearance of the dispersion.

One of the principal problems associated with emulsions is their thermodynamic instability. Emulsions are

dispersed systems that consist of two, completely or partially, immiscible liquids with one liquid being dispersed in the other in the form of droplets (4). The presence of an interface at which polar and nonpolar molecules are in direct contact with each other increases the overall free energy of the system. This increase is directly proportional to the interfacial area. The system has the tendency to decrease the interfacial area to minimize the unfavorable interactions between dispersed and continuous phase molecules. As a consequence, emulsions tend to break down over time.

There are a number of mass transport mechanisms by which this breakdown can occur. The principle instability mechanisms in emulsions are gravitational separation, flocculation, coalescence, and Ostwald ripening (8–11). Gravitational separation occurs if there is a density difference between the continuous and dispersed phases. This causes particles to either cream or sediment. Flocculation is the aggregation of individual oil droplets to form loose agglomerates of fractal structures while retaining their individuality. Coalescence is the merging of two droplets to form one larger droplet through collision of two individual droplets.

Ostwald ripening is the process whereby larger droplets grow at the expense of smaller ones, because of the transport of dispersed phase molecules from the smaller to the larger droplets through the intervening continuous phase (12–14). The driving force for this process is the increased solubility of dispersed phase molecules in the continuous phase for smaller droplets (15). In a polydisperse system, the concentration of dispersed phase molecules that are dissolved in the bulk phase and are located within the vicinity of smaller droplets is higher than the concentration the surrounds larger droplets. As a result of this concentration gradient there is a net movement of dispersed phase mol-

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ecules from smaller to larger droplets. Ostwald ripening can dominate the overall stability of an emulsion if the solubility of the dispersed phase in the continuous phase is appreciable; that is, emulsion droplets consisting of small molecular weight aromatic compounds can double their radius in a matter of a few hours (16). As a consequence, the particle size distribution of the system rapidly changes. Practical examples of food systems that are highly susceptible to Ostwald ripening are beverage emulsions. Because droplet size and concentration directly affect the bulk physicochemical properties of emulsions such as their interaction with light, it can be expected that the appearance and color of an emulsion will change as a result of Ostwald ripening.

The purpose of the present study was to examine the influence of Ostwald ripening on the spectral reflectance and color of oil-in-water emulsions in the presence and absence of dye. Furthermore, the spectral reflectance and color of emulsions was related to their droplet characteristics using light scattering theory.

MATERIALS AND METHODS

Materials. *n*-Hexadecane (>99% pure), *n*-octadecane (>99% pure), and sodium dodecyl sulfate (SDS) were obtained from Sigma Chemical Co. (St. Louis, MO). A red food dye (FD&C Red No. 40) was obtained from Hilton Davis (Cincinnati, OH). Double-distilled and deionized water was used to prepare all solutions and emulsions.

Emulsion Preparation. A 50 mM SDS solution was used as the aqueous surfactant solution. A coarse premix was prepared by homogenizing 25 wt % *n*-hydrocarbon with the 50 mM surfactant solution in a high-speed blender (Waring Product Division, New Hartford, CT). Emulsion premixes were further homogenized using either a high-pressure valve homogenizer (APV Gaulin, Limited, West Sussex, U.K.) or a sonicator (B. Braun Biotech., Melsungen, Germany) to obtain a range of droplet radii between 0.05 and 1.5 μm . Emulsions were then diluted with 50 mM surfactant solution to give final concentrations of 5 wt % oil droplets and either 0 or 0.025% Red No. 40 dye. All emulsions were prepared and stored at $25 \pm 2^\circ\text{C}$ for a period of 2 weeks, and the droplet size and optical properties were analyzed at regular intervals.

Droplet Size Characterization. A laser diffraction instrument (Horiba LA-900, Horiba Instruments Inc., Irvine, CA) was used to measure the droplet size of emulsions containing particles with $r > 0.1 \mu\text{m}$. A relative refractive index of 1.08 (= refractive index of oil/refractive index of aqueous phase) was used by the instrument to calculate the droplet size distribution. Droplet size measurements are reported as the mean diameter $\bar{r} = \sum n_i r_i / \sum n_i$, where n_i is the number of droplets with radius r_i . A number of emulsions contained droplets that were too small to analyze using the laser diffraction instrument. The mean droplet size of these emulsions was determined using a turbidimetric technique (3). Emulsions were diluted with distilled water and placed in a quartz cuvette, and their turbidity was measured at 800 nm using a UV-vis spectrophotometer (UV-2101PC, Shimadzu Scientific Instruments, Columbia, MD). The mean droplet size was then estimated using the equation (3)

$$\bar{r} = \bar{r}_0 \sqrt[3]{\tau_0 / \tau} \quad (1)$$

where \bar{r} and τ are, respectively, the mean radius and turbidity of the unknown emulsion and \bar{r}_0 and τ_0 are, respectively, the radius and turbidity of an emulsion of known droplet size (the emulsion with the smallest droplets that could be measured using the laser diffraction technique). Equation 1 is valid for droplets with sizes much smaller than the wavelength of light ($\bar{r} < \lambda/2\pi$); that is, the size of emulsion droplets at wavelength of 700 nm emulsions must be smaller than ~ 100 nm to

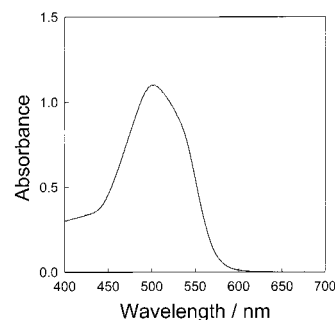


Figure 1. Absorbance spectrum of 0.025 wt % red dye solution (FD&C Red No. 40).

accurately determine their radius. The largest mean radius analyzed with this method was 100 nm, which was the limit of the laser light scattering analyzer.

For both laser diffraction and turbidity measurements the emulsions were diluted to 0.01 wt % with 50 mM SDS solution prior to analysis to avoid multiple scattering effects.

Absorbance and Spectral Reflectance Measurements. Absorbance of dye solutions and spectral reflectance of emulsions were measured with a UV-vis spectrophotometer (UV-2101PC, Shimadzu Scientific Instruments). During the measurements emulsions were contained in quartz cuvettes with a 1 cm path length. Spectra were obtained over the wavelength range of 380–780 nm using a scanning speed of 200 nm min^{-1} . Absorbance measurements were made using a standard double-beam arrangement, with the absorption of the dye solution being measured relative to that of a reference cell containing distilled water. Spectral reflectance measurements were made using an integrating sphere arrangement (ISR-260, Shimadzu Scientific Instruments). The spectral reflectance of the emulsions was measured relative to a barium sulfate (BaSO_4) standard white plate.

Color Measurements. The color of the emulsions was measured using an instrumental colorimeter (Labscan II, Hunter Associates Laboratory, Reston, VA), which was calibrated using a white color standard tile with tristimulus values $X = 78.54$, $Y = 83.18$, and $Z = 85.80$ (Standard LS-13556, Hunter Associates Laboratory). Daylight (D_{65}) was used as a standardized light source. A fixed amount of emulsion sample was poured into the measurement cup, which was then surrounded with a black paper strip and covered with a white tile before the measurement was carried out. All samples were briefly shaken prior to measurement to ensure uniform distribution of emulsion droplets throughout the system and to prevent accumulation or depletion of oil droplets at the scattering interface. The instrument provides the color of the samples in terms of the L , a , and b color space system (17). In this color space, L represents the "lightness" and a and b are color coordinates: $+a$ is the red direction, $-a$ is the green direction, $+b$ is the yellow direction, and $-b$ is the blue direction (18, 19).

RESULTS AND DISCUSSION

Absorbance of Dye Solution. The absorption spectrum of a 0.025 wt % solution of the red dye used in the experiments is shown in Figure 1. The red dye had a single broad absorption peak at $\lambda_{\text{max}} \approx 500$ nm. As shown by Weiss and Liao (7), the absorption at λ_{max} is directly proportional to the dye concentration for solutions containing ≤ 0.03 wt % dye. The solution containing 0.025 wt % dye had a maximum absorption of 1.1 and was chosen as standard dye concentration for all subsequent experiments.

Influence of Droplet Characteristics on Optical Properties of Emulsions. Before carrying out the Ostwald ripening experiments it was necessary to

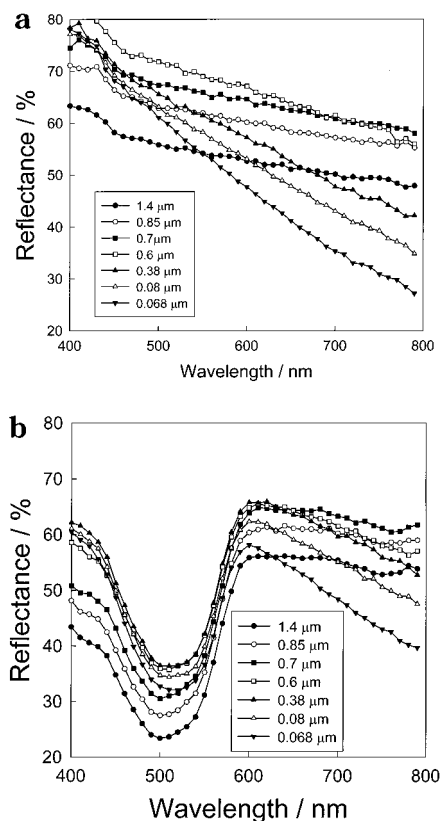


Figure 2. Reflectance spectra of 5 wt % *n*-octadecane-in-water emulsions containing mean droplet sizes ranging from 0.05 to 1.5 μm (a) without red dye and (b) containing 0.025 wt % red dye.

examine the influence of droplet size on the spectral reflectance and color of hydrocarbon oil-in-water emulsions. The spectral reflectance of a series of 5.0 wt % *n*-octadecane oil-in-water emulsions containing a range of mean droplet diameters (0.05–1.5 μm) was measured (Figure 2). Droplet size had a pronounced influence on the spectral reflectance for both the dye-free and dye-containing emulsions. In the absence of dye, the spectral reflectance decreased with increasing wavelength for all droplet sizes (Figure 2a). In addition, at high wavelengths the reflectance increased with droplet size up to a radius of ~0.5 μm, after which it decreased. This maximum is observed because droplets have an optimum scattering efficiency that occurs when their radius is approximately equal to the wavelength of light (1, 3). In the presence of dye, the spectral reflectance had a trough between 380 and 600 nm, which corresponded to the peak in the absorption spectra of the dye (Figure 1). This trough occurs because light is selectively absorbed by the emulsion at these wavelengths and therefore is not reflected. At wavelengths >600 nm the emulsion containing dye exhibited spectral characteristics similar to those of the dye-free emulsion.

The color *L*, *a*, and *b* values as influenced by droplet size for 5 wt % *n*-octadecane oil-in-water emulsions are presented in Figure 3. Emulsion "lightness" (*L* value) initially increased with droplet radius up to about $r = 0.5 \mu\text{m}$, after which it decreased (Figure 3a). The maximum in the *L* value occurs when the droplets have a radius that corresponds to their greatest scattering efficiency (3). The *L* value of the dye-containing emulsions was lower than that of the dye-free emulsions, as has been observed in previous studies (5). Emulsion chromaticity (*a* and *b* values) was also influenced by

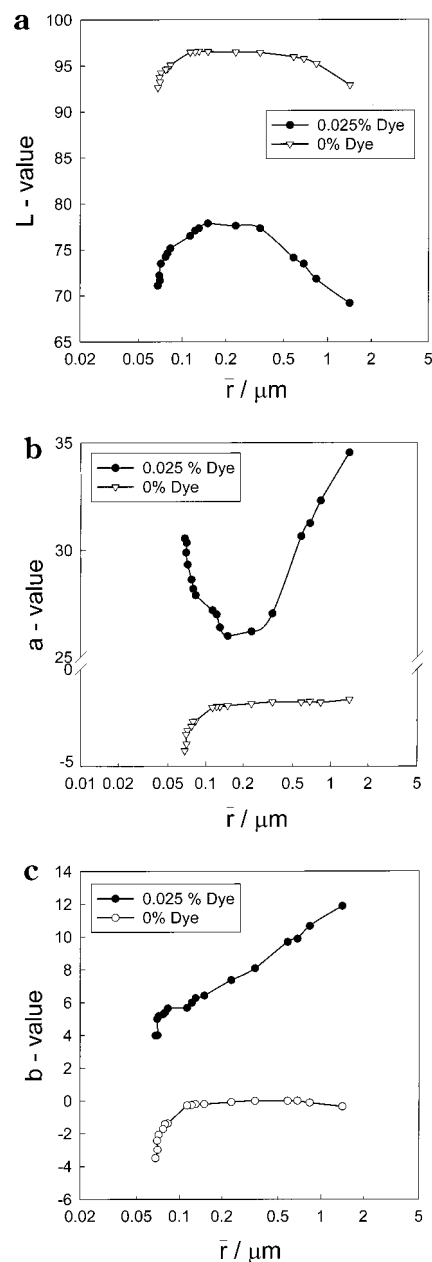


Figure 3. Tristimulus coordinates [(a) *L* value, (b) *a* value, and (c) *b* value] of 5 wt % *n*-octadecane-in-water emulsions as a function of mean droplet radius. Emulsions contain either 0 wt % red dye or 0.025 wt % red dye.

droplet size. In the absence of dye, the *a* and *b* values tended toward zero as the droplet size increased up to ~0.2 μm, after which it remained fairly constant (Figure 3b,c). The slightly negative *a* and *b* values at small droplet sizes indicate that the emulsions had a blue-green color, whereas the zero values at larger droplet sizes indicate that they appeared white. In the presence of dye, the *a* value decreased as the droplet size increased to ~0.2 μm, after which it increased, indicating that the emulsion first became less red and then more red as the droplet size increased. The *b* value of the dye-containing emulsions increased as the droplet size increased, indicating that the emulsions became progressively more yellowish in appearance. The blue color of emulsions containing very small emulsion droplets can be explained in terms of light scattering. The interaction of very small emulsion droplets with light at wavelengths above the droplet size is strongly

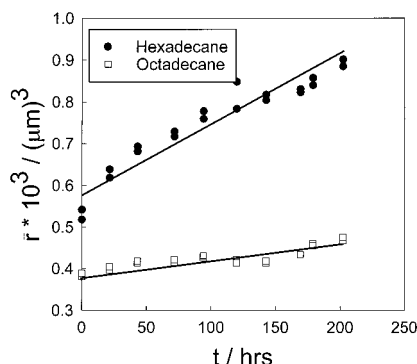


Figure 4. Cube of mean droplet radius as a function of aging time for oil-in-water emulsions containing either 5 wt % *n*-hexadecane or 5 wt % *n*-octadecane.

reduced, and as a consequence light is no longer scattered by the particles. Longer wavelengths in the red and yellow spectra pass the emulsion undisturbed, whereas light waves with smaller wavelengths in the blue-purple region are still scattered, giving the entire emulsion a blue color. At larger droplet sizes, light is scattered in the entire region of the visible spectra, giving the emulsions a white color.

These results confirm that the optical properties of emulsions depend on droplet size. Any physicochemical process that alters the droplet size should change their appearance.

Influence of Ostwald Ripening on Droplet Size.

The evolution of the mean droplet size of 5 wt % *n*-hexadecane and *n*-octadecane oil-in-water emulsions was monitored using laser diffraction and turbidity measurements. The droplet size of the *n*-hexadecane emulsion increased significantly over time, whereas that of the *n*-octadecane emulsion increased by only a few nanometers. Figure 4 shows a plot of the cube of the mean droplet size of the *n*-hexadecane and *n*-octadecane emulsions as a function of storage time. Figure 4 can be used to calculate the rate of Ostwald ripening. The rate of Ostwald ripening in a dilute emulsion is given by the equation (12, 13):

$$\bar{r}_t^3 - \bar{r}_{t=0}^3 = 8\gamma D c_{r \rightarrow \infty} V_m^2 t / 9RT \quad (2)$$

where \bar{r} is the mean droplet radius, t is the time, γ is the interfacial tension at the oil-water interface, D is the diffusion coefficient of the oil through the aqueous phase, $c_{r \rightarrow \infty}$ is the solubility of the oil (when contained in an infinitely large droplet) in the aqueous phase, V_m is the molar volume of the oil, R is the gas constant, and T is the absolute temperature. The main reason that the Ostwald ripening rate of the *n*-hexadecane droplets is appreciably greater than that of the *n*-octadecane droplets is because hexadecane has a higher water solubility (20, 21). Equation 2 indicates that the cube of the mean droplet size of the emulsions should increase linearly with time, with a slope that depends on the physical characteristics of the oil and aqueous phases. There was an approximately linear relationship between \bar{r}^3 and time for the emulsions used in this study (Figure 4), with slopes of $1.6 \times 10^{-3} \mu\text{m}^3 \text{h}^{-1}$ for the *n*-hexadecane droplets and $0.3 \times 10^{-3} \mu\text{m}^3 \text{h}^{-1}$ for the *n*-octadecane droplets.

Influence of Ostwald Ripening on Optical Properties of Emulsions. The spectral reflectance of 5 wt % *n*-hexadecane and *n*-octadecane oil-in-water emulsions was measured over 202 h. In the absence of dye

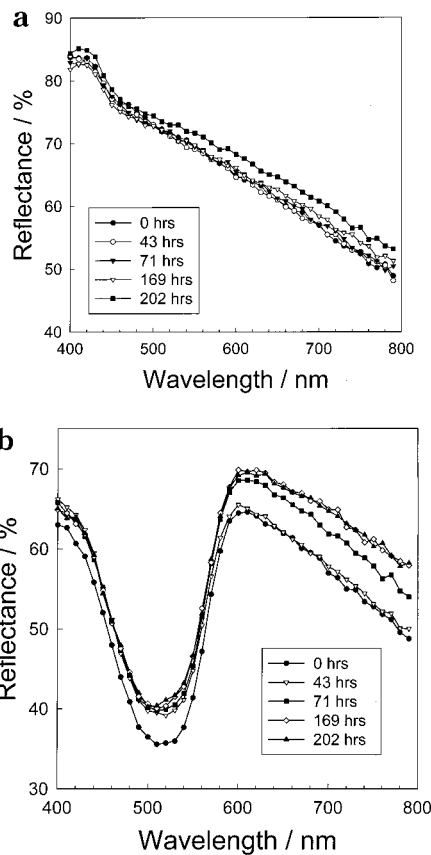


Figure 5. Reflectance spectra of 5 wt % *n*-hexadecane-in-water emulsions during aging of (a) emulsion without dye and (b) emulsion with 0.025 wt % red dye.

the spectral reflectance of the *n*-hexadecane emulsion decreased with increasing wavelength (Figure 5a). In the presence of dye, there was a trough at wavelengths corresponding to the absorbance peak of the red dye (Figure 5b). In the *n*-hexadecane emulsion, there was also a significant increase in the spectral reflectance with time due to the growth of the droplets. No visible change in the spectral reflectance of the *n*-octadecane emulsion was observed during the same time (data not shown). This was because the rate of Ostwald ripening in the *n*-octadecane emulsion was ~ 5 times smaller than the rate of Ostwald ripening in the *n*-hexadecane emulsions.

The L , a , and b color coordinates of 5 wt % *n*-hexadecane and *n*-octadecane oil-in-water emulsions during the time of storage were measured as well (Figure 6). The "lightness" (L value) of the *n*-hexadecane emulsion increased, the a value decreased, and the b value increased during the course of the experiment. These changes occurred because the droplet radius increased from about 0.05 to 0.3 μm during the course of the experiment. If the droplets had grown any larger, we would have expected the lightness to start to decrease and the a value to increase. There were only slight alterations in the L , a , and b values of the *n*-octadecane emulsion because there was little change in the droplet size during the experiment.

The fact that the tristimulus color coordinates were sensitive to changes in droplet size suggests that colorimetry measurements could be used to nondestructively monitor the kinetic stability of concentrated emulsions. There was a pronounced change in the L , a , and b values with droplet size for emulsions containing

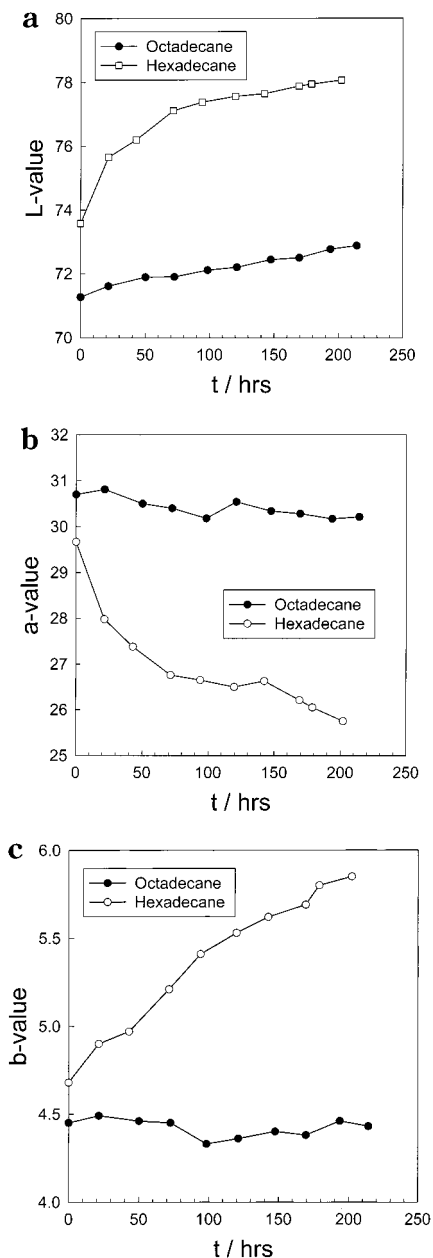


Figure 6. Tristimulus coordinates [(a) *L* value, (b) *a* value, and (c) *b* value] of *n*-hexadecane and *n*-octadecane emulsions with 0.025 wt % red dye during aging at 25 °C.

small droplets (<0.2 μm), although there was little change at larger sizes. Colorimetry may be useful for monitoring droplet size changes due to Ostwald ripening or coalescence in emulsions containing small droplets, such as parenteral emulsions (23).

Light Scattering Theory. As shown by McClements (22) and Weiss and Liao (7), the color and appearance of emulsions can be explained in terms of light scattering theory. A light beam that propagates through a random medium containing particles capable of scattering and absorbing part of the electromagnetic wave will be attenuated after traversing a distance *dl*. The change in intensity *I* of the electromagnetic wave is given as

$$dI = N\pi r^2 Q I dl \quad (3)$$

where *N* is the number concentration of particles, *r* their radius, and *Q* the extinction coefficient. The extinction

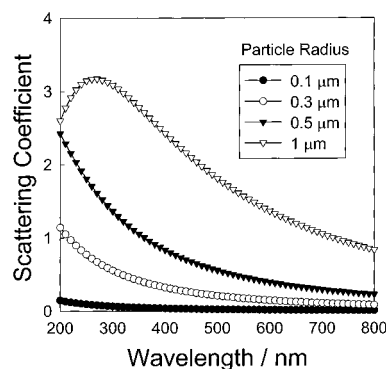


Figure 7. Scattering efficiency *Q* as a function of wavelength calculated from eq 4 using a relative refractive index of 1.08 and sizes ranging from 0.1 to 5 μm.

coefficient for partially absorbing spheres with a relative refractive index close to one is given as (1)

$$Q = 2 - 4 e^{-\rho \tan \beta} (\cos \beta / \rho) \sin(\rho - \beta) - 4 e^{-\rho \tan \beta} (\cos \beta / \rho)^2 \cos(\rho - 2\beta) + 4 (\cos \beta / \rho)^2 \cos 2\beta \quad (4)$$

where ρ is the real parameter

$$\rho = 2x(n_{\text{Re}} - 1) \quad (5)$$

and $x = 2\pi r / \lambda$ is a size parameter and n_{Re} is the real part of the index of refraction. The parameter β assumes values between 0 and ∞ and is defined as

$$\tan \beta = n_{\text{Im}} / (n_{\text{Re}} - 1) \quad (6)$$

where n_{Im} is the imaginary part of the index of refraction. In a first approximation, emulsion droplets are assumed to be nonabsorbing spheres with β being equal to zero. On the basis of eq 4 we can now explain the results that were obtained in the experiments. Figure 7 shows a plot of the extinction coefficient *Q* as a function of the wavelength of incident light of emulsions with droplet mean radii of 0.1, 0.3, 0.5, and 1 μm. The extinction coefficient *Q* was calculated from eq 4 using a relative refractive index of 1.08, which corresponds to the ratio of the refractive index of *n*-hexadecane to the refractive index of water at 25 °C. The scattering coefficient *Q* decreases strongly over the entire wavelength spectra as the droplet size of emulsions decreases. This indicates that scattering is lower as the droplet size is smaller; thus, the emulsion begins to appear transparent. The influence of the droplet size on the scattering coefficient *Q* was calculated using a constant wavelength of 650 nm (Figure 8). The wavelength was chosen because it corresponds approximately to the wavelength of the He-Ne laser used in the static laser light scattering experiments. We can see that as the droplet size increases, the scattering coefficient increases to reach a maximum at ~2 μm. Above 2 μm, the scattering coefficient follows the pattern of a damped sinusoidal oscillation.

Figure 8 illustrates two important facts that are of direct consequence to this research: (1) The influence of Ostwald ripening on the color and appearance of the emulsion is strong if the initial emulsion consists of very small droplets, that is, with sizes below 0.1 μm. (2) The measurement of the scattering coefficient at a single wavelength cannot be used to predict particle sizes of large droplets because the value of the scattering coefficient is the same for several particle sizes; that is,

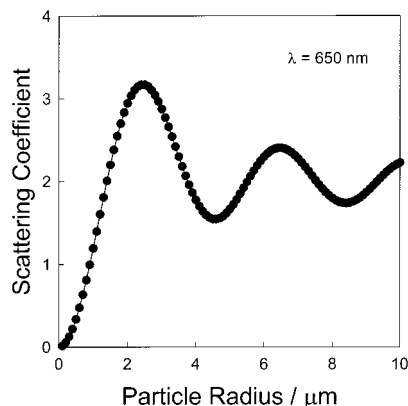


Figure 8. Scattering efficiency Q as a function of droplet radius (at $\lambda = 650$ nm and $n = 1.08$).

there exists no single solution for Q . It should be noted, though, that the spectral dependence of the scattering coefficient is specific enough to predict particle sizes and thus confirms our hypothesis that colorimetry can be used to monitor droplet size changes in emulsions.

CONCLUSIONS

The optical properties of emulsions were influenced by Ostwald ripening because the transport of oil molecules from smaller to larger droplets caused a gradual increase in the mean droplet size. Thus, Ostwald ripening not only reduces the stability of the emulsion but also alters the color and appearance of the product. Changes in droplet appearance during storage are undesirable in a number of products, such as cosmetics, foods, beverages, and pharmaceuticals. Food manufacturers therefore need to be able to predict these changes to determine the shelf life of their products. The research shows that light scattering theory can be used to model color changes in dynamically complex emulsions that exhibit molecular mass transport phenomena such as Ostwald ripening. The presented work has therefore direct practical implications for the food industry. Changes in emulsion appearance due to Ostwald ripening would be most important for products that contained dispersed phases with a significant solubility in the continuous phase, such as fruit beverages. Our experiments also suggest that colorimetry measurements may be used to nondestructively monitor the stability of emulsions containing small droplets.

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