Influence of Droplet Size and Concentration on the Color of Oil-in-Water Emulsions

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The influence of droplet size $(0.2-15 \ \mu\text{m})$ and concentration (0-20 wt %) on the turbidity and color of *n*-hexadecane oil-in-water emulsions containing a blue food dye has been investigated. Emulsion turbidity was maximum for droplets with diameters of $\sim 2 \ \mu\text{m}$ (at 650 nm). Colorimetry measurements of the tristimulus coordinates (L, a, b) of emulsions indicated that they became less light, more green, and more blue as droplet size increased and more light as droplet concentration increased. Sensory analysis showed emulsions became lighter and less blue as droplet size decreased and droplet concentration increased. The observed behavior can be explained in terms of the scattering and absorption of light by emulsions. Our results show that food emulsion appearance can be optimized by carefully controlling droplet characteristics, such as size and concentration.

Keywords: emulsion; color; turbidity; droplet size; droplet concentration; light scattering

INTRODUCTION

Many natural and manufactured food products exist in the form of oil-in-water emulsions, e.g., milk, cream, fruit beverages, salad dressings, mayonnaise, soups, sauces, and infant formulations (Dickinson and Stainsby, 1982; Friberg, 1980; Dickinson, 1992; McClements, 1998). The quality of emulsion-based food products is largely determined by their appearance, taste, texture, and shelf life. A great deal of research has been carried out by colloid scientists to elucidate the factors which influence the texture and shelf life of food emulsions (Dickinson, 1992; McClements, 1998). Recently, there has been increasing interest in studying flavor distribution and release in food emulsions (McNulty, 1987; Overbosch et al., 1991; Baker and Mela, 1996; Guyot et al., 1996; Landy et al., 1996). Given its important role in determining the quality of food products (Clydesdale 1978, 1993, Hutchings 1994), suprisingly little work has been carried out on the factors which influence emulsion appearance.

The overall appearance of a food is determined by the way that it interacts with radiation in the visible region of the electromagnetic spectrum, e.g., reflection, transmission, absorption, and scattering (Clydesdale, 1978; Hutchings, 1994). These interactions can be extremely complex and are governed by the unique composition and structure of a food. One of the major factors determining the appearance of oil-in-water food emulsions is the presence of oil droplets. The three major characteristics of oil droplets which influence emulsion appearance are their size, concentration, and refractive index (Farinato and Rowell, 1983). The influence of droplet size and concentration on the turbidity (cloudiness) of beverage oil-in-water emulsions has been studied (Hernandez and Baker, 1991; Hernandez et al., 1991; Dickinson, 1994). A maximum in emulsion turbidity was observed when the droplet diameter was

around 1 μ m (at λ = 650 nm). This meant that the oil droplet concentration required to produce a desirable emulsion cloudiness could be minimized by using droplets with diameters around this size. The importance of droplet refractive index on the scattering of light and appearance of emulsions has also been investigated (Walstra, 1968; Tainse et al., 1996). The greater the difference between the refractive index of the droplets and surrounding liquid, the stronger the light scattering, and therefore the more turbid the emulsion. A number of workers have studied the factors which influence the color of food emulsions and other colloidal systems, but none of these studies have specifically focused on the precise role of the particle characteristics (Huang et al., 1970; Gullet et al., 1972). For this reason, we decided to investigate the influence of droplet size and concentration on the color and turbidity of oil-inwater emulsions containing a blue food dye.

PHYSICAL BASIS OF EMULSION COLOR

Ideally, one would like to be able to predict the color of a food emulsion (e.g., its L, a, b tristimulus values) from a knowledge of its composition and structure, e.g., the concentration and type of chromophores and the size, concentration, and nature of the droplets. In practice, it is difficult to do this because of the complexity of the physical processes involved, especially in concentrated emulsions. Nevertheless, a consideration of the interactions between light waves and emulsions can provide some useful insights into the influence of droplet characteristics on emulsion appearance.

Dilute Emulsions. The optical characteristics of dilute emulsions, in which multiple scattering effects are negligible, can be described by the following equation (Heimenz 1986):

$$(I_T/I_0) = \exp(-\alpha \Delta x) \tag{1}$$

where $I_{\rm T}$ is the intensity of transmitted light, I_0 is the intensity of incident light, α is the extinction coefficient,

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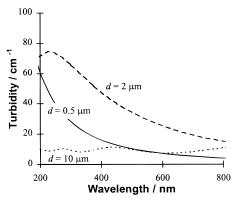


Figure 1. Predicted wavelength dependence of the turbidity of emulsions containing different droplet sizes.

and Δx is the sample thickness. The extinction coefficient contains the influence of both absorption and scattering effects and is the sum of the absorbance (ϵ) and turbidity (τ): $\alpha = \epsilon + \tau$. The extinction coefficient is dependent on the size and concentration of droplets in an emulsion through scattering effects and on the type and concentration of chromophores present through absorption effects:

$$\alpha = N\pi R^2 Q \tag{2}$$

where *N* is the number of droplets per unit volume, *R* is the droplet radius, and *Q* is the efficiency factor, which contains contributions from both scattering and absorption. The efficiency factor depends on the wavelength of light (λ), the droplet radius, the real (*n*) and imaginary (*k*) part of the complex refractive index (*m* = *n* + i*k*), and the angle of observation (ϕ):

$$Q = f(\lambda, R, n, k, \phi) \tag{3}$$

Theories are available to calculate Q for dilute emulsions from a knowledge of the physical properties of the droplets and surrounding medium (Kerker, 1969; Bohren and Gilara, 1979; Hiemenz, 1986). In general, these theories are extremely complicated and can only be solved numerically on a digital computer (Dave, 1969). Nevertheless, much simpler analytical expressions can be derived when certain restrictions are put on the physical characteristics of the system (Kerker, 1969). For example, if one assumes that the wavelength of light is greater than the droplet size ($\lambda < R$), that the refractive index of the droplets is close to that of the surrounding medium ($n_1 \sim n_2$), and that the liquids are nonabsorbing (Walstra, 1964),

$$Q = 2 - \frac{4}{\beta} \sin\beta + \frac{4}{\beta^2} (1 - \cos\beta) \tag{4}$$

$$\beta = \frac{4\pi Rn}{\lambda} |n-1| \tag{5}$$

Turbidity spectra predicted by using eqs 2 and 4 are shown in Figure 1 for three oil-in-water emulsions with different droplet sizes. These predictions clearly show that droplet size has a pronounced influence on the wavelengths of light which are capable of being transmitted through a dilute emulsion. When certain wavelengths are selectively removed from the light beam the color of an emulsion will change, even in the absence of a dye (Hiemenz, 1986). When a dye is present, the overall color of an emulsion is determined by a complex interplay between absorption and scattering effects (Bohren and Gilara, 1979).

Concentrated Emulsions. The optical characteristics of concentrated emulsions are much more difficult to predict than those of dilute emulsions because of multiple scattering effects (Kerker, 1969; Bailey and Cannell, 1994). Usually, it is not possible to transmit a light beam through a concentrated emulsion because of the high degree of attenuation caused by droplet scattering. As a consequence, the appearance of a concentrated emulsion is mainly determined by that portion of the light which is "reflected" from its surface (or close to the surface). When a light wave is incident on the surface of a concentrated emulsion, part of it is reflected at the surface, while the rest is transmitted into the emulsion. As the transmitted wave propagates through the emulsion it may be absorbed by any chromophores or scattered by any droplets. The light returning from the emulsion is therefore the result of reflected, transmitted, scattered, and absorbed light. The degree of absorption and scattering which occurs depends on the wavelength of light. Consequently, when a beam of white light is incident upon an emulsion the intensity and color of the light which returns depends on the degree of scattering and absorption by the emulsion, which in turn depends on the droplet characteristics (size and concentration) and chromophore characteristics (absorption spectra and concentration). In principle, it should be possible to predict the properties of light reflected from a concentrated emulsion using eqs 1-3. In practice, this is not usually possible because these equations do not take into account the effects of multiple scattering, which are important in concentrated systems. It is therefore necessary to adopt a different theoretical approach to describe the optical properties of concentrated emulsions.

The optical characteristics of concentrated emulsions can be described by the Kubelka–Munk theory (Francis and Clydesdale, 1975; Hutchins, 1994). This theory enables one to determine the relative contribution of scattering and absorption to the reflection of a light wave from a concentrated emulsion:

$$F_{\rm KM} = \frac{K}{S} = \frac{(1 - R_{\odot})^2}{2R_{\odot}}$$
(6)

where $F_{\rm KM}$ is the Kubelka–Munk parameter, K and S are the absorption and scattering coefficients, and R_{∞} is the reflectance from an infinitely thick sample at a given wavelength. The scattering coefficient is proportional to the droplet concentration, while the absorption coefficient is proportional to the chromophore concentration. When an emulsion is highly colored, the absorption of light dominates (high $F_{\rm KM}$) and the reflectance is low. As scattering effects become more important (low $F_{\rm KM}$), the reflectance increases and the emulsion appears "lighter" in color. We would therefore expect the color of a concentrated emulsion to become lighter as the efficiency of light scattering by the droplets increased.

MATERIALS AND METHODS

Materials. *n*-Hexadecane (>99% pure) was obtained from Sigma Chemical Company (St. Louis, MO). Xanthan, a polysaccharide derived from *Xanthomonas campestris*, was obtained from Kelco International (London, UK). Blue food coloring (FD&C blue #1, Durkee, Burns Philp Food Inc,. San Francisco, CA) was obtained from a local supermarket. A 0.1 vol % stock solution of this dye was prepared by dilution with distilled water. Double distilled and deionized water was used to prepare all solutions and emulsions.

Emulsion Preparation. *Effect of Droplet Size.* In one series of experiments we examined the influence of droplet size on emulsion turbidity and color. For this reason, we prepared a series of emulsions with similar final droplet concentrations but different mean droplet diameters.

Emulsions containing relatively large droplet sizes (>1 μ m) were prepared by homogenizing 40 wt % n-hexadecane and 60 wt % surfactant solution (2 wt % Tween 20 and 0.5 wt % xanthan in water) in a high-speed blender (model 38BL54, Warring Products Division, New Hartford, CT). Different mean droplet sizes were produced by blending the emulsions for different times (5-80 s). These emulsions were then diluted to a droplet concentration of 10 wt % n-hexadecane with surfactant solution (0.5 wt % Tween 20 and 0.5 wt % xanthan in water). Emulsions containing relatively small droplet sizes (<1 μ m) were prepared by homogenizing 10 wt % \hat{n} -hexadecane and 90 wt % surfactant solution (0.5 wt % Tween 20 in water) in a high-pressure valve homogenizer (Rannie 8.30R, Wilmington, MA). Emulsions with different mean droplet diameters were produced by passing them through the homogenizer at different pressures and for varying numbers of passes. Powdered xanthan was then added to the emulsions containing small droplets to give a final concentration of 0.5 wt % in the aqueous phase.

For the coarse emulsions, xanthan was added to the aqueous phase before homogenization to prevent droplets from rapidly creaming and coalescing during and immediately after blending. For the fine emulsions, xanthan was added after homogenization because otherwise it would have been difficult to pass the highly viscous samples through the high-pressure valve homogenizer.

For turbidity measurements, the 10 wt % emulsions were diluted to a final droplet concentration of 0.005 wt % *n*-hexadecane with distilled water. For color measurements, the 10 wt % emulsions were diluted with dye solution to give a series of colored emulsions with different mean droplet diameters but constant emulsion composition (9.5 wt % *n*-hexadecane, 0.45% Tween 20, 0.45% xanthan, and 0.005 wt % dye).

Effect of Droplet Concentration. In another series of experiments we examined the influence of droplet concentration on emulsion color. We therefore prepared emulsions with constant droplet sizes but different droplet concentrations. The effect of concentration on emulsions with three different droplet sizes was studied: small ($\sim 0.3 \,\mu m$), medium ($\sim 2 \,\mu m$), and large (~10 μ m). Oil-in-water emulsions were prepared with a final composition of 20 wt % n-hexadecane and 80 wt % surfactant solution (1 wt % Tween 20 and 0.5 wt % xanthan in water). The emulsions containing small droplets were prepared by using the high-pressure valve homogenizer, while those containing medium or large droplets were prepared by using the high-speed blender. Emulsions with a range of droplet concentrations (0-20 wt %) were prepared by mixing different ratios of 20 wt % emulsion and 0.5 wt % aqueous xanthan solution. A constant amount of the 0.1 vol % dye solution was added to each of the emulsions to give a final dye concentration of 0.005 wt %.

Droplet Size Measurements. A static light scattering technique (Horiba LA-900, Horiba Instruments Incorporated, Irving, CA) was used to measure the droplet size distribution of emulsions. This instrument measures the angular dependence of the scattered light intensity when a laser beam passes through a dilute emulsion. The intensity vs angle profile is then converted to a droplet size distribution by the instrument using Mie theory. 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. To avoid multiple scattering effects emulsions were diluted with distilled water prior to analysis so that the final droplet concentration was \sim 0.005 wt %.

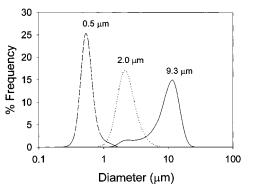


Figure 2. Selected droplet size distributions of emulsions measured by using a light scattering technique.

Most droplet size measurements are reported as the volume– surface mean diameter: $d_{32} = \sum n_i d_i^{3/} \sum n_i d_i^{2}$, where n_i is the number of droplets with diameter d_i . The full droplet size distributions of selected emulsions are shown in Figure 2. The droplet size distribution of the emulsions was measured at the beginning and end of the experiments and was found to remain constant, which indicated no coalescence or Ostwald ripening occurred during the experiments. All measurements were carried out before the dye was added to the emulsion to avoid complications in the droplet size analysis associated with the wavelength-dependent absorption of the dye.

Absorbance and Turbidity Measurements. Absorbance and turbidity spectra of dye solutions and emulsions were measured by using a UV–visible spectrophotometer (UV-2101PC, Shimadzu Scientific Instruments, Columbia, MD). Spectra were obtained over the wavelength range 200–800 nm, using a scanning speed of 700 nm min⁻¹.

Color Measurement. The color of the emulsions was measured on an instrumental colorimeter (Labscan II, Hunter Associates Laboratory, Reston, VA), which was calibrated by using a white color standard tile with tristimulus values: X= 78.54, Y = 83.18, and Z = 85.80 (Standard No. LS-13556, Hunter Associates Laboratory, Reston, VA). A fixed amount of emulsion sample was poured into the measurement cell, which was then covered with the white tile before the measurement was carried out. The instrument reports the color of the samples in terms of the *L*,*a*,*b* color space system (Figure 3). In this color space, *L* represents the lightness and *a* and *b* are color coordinates: where +a is the red direction, -a is the green direction, +b is the yellow direction, and -bis the blue direction (Francis and Clydesdale, 1975). The data are also represented in terms of hue angle (H), chroma (C), and color difference (ΔE) to highlight comparative differences between samples:

$$H = \tan^{-1}\frac{b}{a} \tag{7}$$

$$C = (a^2 + b^2)^{1/2}$$
(8)

$$\Delta E = (\Delta L^2 + \Delta a^2 + \Delta b^2)^{1/2} \tag{9}$$

where ΔL , Δa , and Δb are the differences in the specified tristimulus coordinate between the sample and a reference. The hue angle provides a quantitative measure of the change in the hue of a material, i.e., the attribute of color perception which is denoted by the terms "blue", "green", "yellow", "red", etc. (Wyszecki and Stiles, 1967).

Sensory. Sensory analysis of emulsion appearance was performed by five untrained panel members (graduate students in the Department of Food Science, University of Massachusetts). Emulsions were poured into glass test tubes (height = 100 mm, internal diameter = 15 mm) and randomly placed in a test tube rack before being presented to the panelists for visual assessment. Panelists were then in-

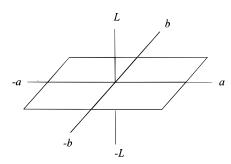


Figure 3. Schematic representation of the *L*,*a*,*b* color space.

Table 1. Panelist Rankings of the Lightness and Blueness of Emulsions with Different Diameters but Same Droplet Concentration (9.5 wt %) and Dye Concentration (0.005 wt %)^a

	mean droplet diameter, μ m							
appearance	0.38	0.5	2.1	5.0	10	30		
lightest bluest	1.5 (0.3) 5.3 (0.3)	1.5 (0.3) 5.7 (0.3)	3 (0) 4 (0)	4 (0) 3 (0)	5 (0) 2 (0)	6 (0) 1 (0)		

 a 1 = lightest or bluest, and 6 = darkest or least blue. Values are given as the mean of five measurements, with the standard deviation in parentheses.

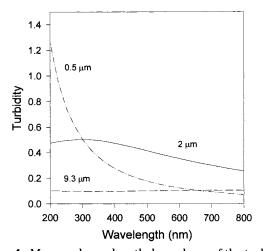


Figure 4. Measured wavelength dependence of the turbidity of 0.05 wt % *n*-hexadecane oil-in-water emulsions containing different droplet sizes.

structed to rank emulsion samples in order of "lightness" and "blueness" on a scale of 1 to 6 (Table 1).

RESULTS AND DISCUSSION

Turbidity of Dye-Free Emulsions. The turbidity of 0.005 wt % n-hexadecane oil-in-water emulsions containing a range of different mean droplet diameters $(d_{32} = 0.2 - 15 \,\mu\text{m})$ was measured in the absence of dye. Turbidity spectra of three emulsions with different mean droplet sizes are presented in Figure 4. These results clearly show that emulsions with the same droplet concentration, but different droplets sizes, have dramatically different turbidity spectra. For example, the emulsion containing the largest droplets had a relatively low turbidity across the whole wavelength range, whereas the one containing the smallest droplets scattered light strongly at the lower wavelengths. The experimental turbidity spectra are in reasonable agreement with those predicted by eqs 1-5 (Figure 1). The major discrepancy between the measured and predicted values was for the 2- μ m droplets. The most likely reason for this discrepancy is that the emulsion turbid-

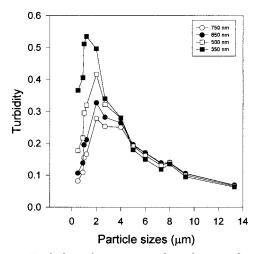


Figure 5. Turbidity of 0.05 wt % *n*-hexadecane oil-in-water emulsions containing different droplet sizes.

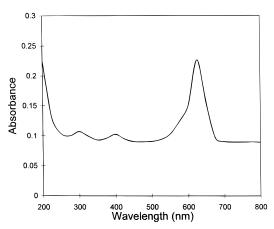


Figure 6. Absorption spectra of 0.05 wt % blue dye solution.

ity is particularly sensitive to d_{32} in this droplet size range, there being a maximum in turbidity at $d_{32} \approx 2$ μ m (Figure 5). In addition, the predicted values were calculated assuming all the droplets had the same size, whereas in reality they have a range of different sizes (Figure 2). Droplet polydispersity therefore has the effect of smoothing out the curves and reducing the height of the turbidity maximum (Hernandez and Baker, 1991).

The experimental data shown in Figure 5 are in good agreement with that of Hernandez and Baker (1991), who also studied the influence of droplet size on emulsion turbidity spectra. Differences in the turbidity spectra of emulsions with the same overall composition indicate differences in the scattering efficiency of the droplets. The scattering efficiency is related to the scattering term (S) in the Kubelka–Munk theory (Hutchings, 1994). One would therefore expect the size of the droplets in a concentrated emulsion to effect its lightness and color because of their influence on the wavelength dependence of the K/S parameter. Our laboratory is currently examining the possibility of predicting the color of emulsions from the Kubelka–Munk theory. To do this it will be necessary to develop a mathematical expression for K/S which depends on the characteristics of the droplets and dye.

Absorbance of Droplet-Free Dye Solutions. Absorbance vs wavelength spectra of aqueous solutions of the blue dye are shown in Figure 6. There was a large maximum in the absorbance spectra around 630 nm,

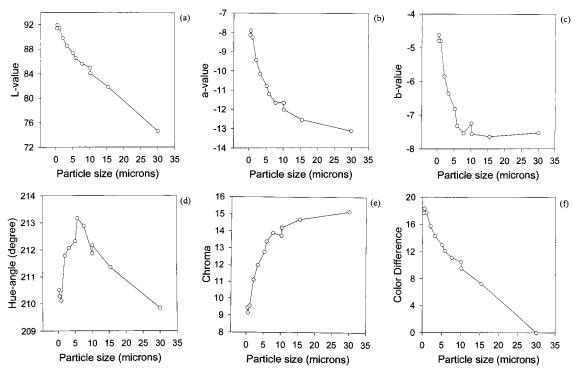


Figure 7. Dependence of *L*, *a*, *b*, color difference, hue angle, and chroma on droplet size for 10 wt % *n*-hexadecane oil-in-water emulsions.

which accounts for the predominantly blue color of the dye. The absorbance of the solutions obeyed the Beer–Lambert law over the concentration range 0-0.01 vol %; i.e., it was linearly dependent on dye concentration (data not shown).

Effect of Droplet Size on Emulsion Color. A series of n-hexadecane oil-in-water emulsions were prepared which had the same droplet concentration (9.5 wt %) and dye content (0.005 wt %) but different droplet diameters ($d_{32} = 0.2 - 15 \,\mu$ m). The color of the emulsions was measured on a colorimeter and presented in the form of the L, a, b tristimulus system and as chroma, hueangle, and color difference (Figure 7). The L value of the emulsions decreased with increasing droplet diameter, while the *a* and *b* values became increasingly negative, indicating the emulsions became darker, more green, and more blue as the droplet size increased. The most likely reason for the increase in the "greeness" and "blueness" of the emulsions with increasing droplet size is that the scattering efficiency of the droplets decreases and therefore the light beam can penetrate further into the emulsion, which increases the absorption.

The color difference of the emulsions (relative to the emulsion containing the largest droplets) became increasingly small as the droplet diameter increased (Figure 7f). The hue-angle had a maximum at a droplet diameter of $\sim 5 \ \mu$ m, while the chroma increased with increasing droplet size (Figure 7e,f). The increase in chroma indicates that the color of the emulsions became more intense as the droplet size increased. The reason for the maximum in the hue-angle is unknown, although the changes in hue-angle are relatively small.

Differences in emulsion appearance due to differences in droplet size could clearly be distinguished by our untrained panel (Table 1). Panelists found an increasing trend in "blueness" and decreasing trend in "lightness" as the droplet size increased. The only exception to this trend was for the two smallest emulsions, where some panelists ranked the 0.38-µm emulsion as being less light and more blue than the 0.5 μ m emulsion. This is probably related to the maximum which occurs in the efficiency of the light scattering at a particular droplet diameter. In our turbidity experiments a maximum occurred at a droplet diameter of $\sim 2 \,\mu m$ (Figure 5). The turbidity of an emulsion is determined by measuring the intensity of light which travels directly through it. Other workers have shown that the maximum scattering efficiency occurs at lower droplet diameters when the light intensity is measured at other angles (Hernandez et al., 1991). The appearance of a concentrated emulsion is determined principally by the light which is "reflected" from its surface, and so the maximum in the scattering efficiency will occur at droplet sizes somewhere below the 2 μ m found from the turbidity measurements. Indeed, there appears to be a maximum in the lightness of the emulsions around a droplet size of about 0.2 μ m (Figure 7a). The existence of a maximum in the scattering efficiency of the reflected light would explain why all of the panelists did not report a progressive trend in the appearance of emulsions with increasing droplet size (Table 1). In addition, the fact that the scattering efficiency has a maximum value at lower droplet sizes for reflected than for transmitted light would account for the fact that no maximum was observed in the L value for droplets around 2 μ m, even though there was a maximum in the turbidity.

Linear regression analysis showed a strong correlation between *L* value and droplet size (r = -0.994), between *L* value and panelist score of "lightness" (r = -0.905), and between *b* value and panelist score of "blueness" (r = 0.978) and a reasonable correlation between *b* value and droplet size (r = 0.736).

Effect of Droplet Concentration on Emulsion Color. Three series of *n*-hexadecane oil-in-water emulsions were prepared which had different droplet concentrations (0–20 wt %) but the same mean droplet diameter ($d_{32} = 0.3$, 2.0, or 10 μ m) and dye content

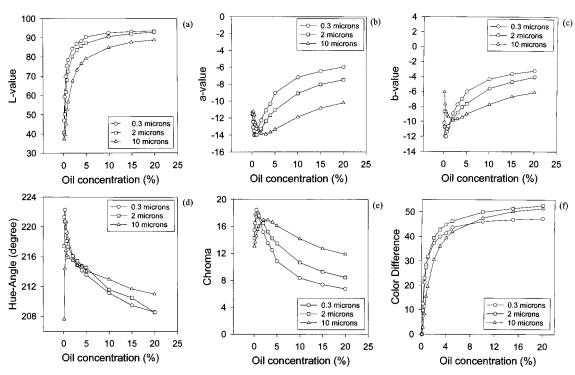


Figure 8. Dependence of *L*, *a*, *b*, color difference, hue angle, and chroma on droplet concentration for *n*-hexadecane oil-in-water emulsions.

Table 2. Compilation of Panelists Rankings of the "Lightness" and "Blueness" of Emulsions with Different Droplet Concentrations but the Same Droplet Diameters ($d_{32} = 0.3 \mu m$) and Dye Concentration (0.005 wt %)^a

appearance	droplet concentration, wt %								
	0.1	0.5	1	5	10	15	20		
lightest bluest	6.8 (0.3) 1 (0)	6.2 (0.3) 2 (0)	5 (0) 3 (0)	4 (0) 4 (0)	2.6 (0.3) 5 (0)	2.4 (0.3) 6 (0)	1 (0) 7 (0)		

a 1 = lightest or bluest, and 6 = darkest or least blue. Values are given as the mean of five measurements, with the standard deviation in parentheses.

(0.005 wt %). The color of the emulsions was measured using a colorimeter and presented in the form of *L*,*a*,*b* tristimulus values and as *chroma*, *hue-angle*, *and color difference* (Figure 8). The *L* value of the emulsions increased steeply with increasing droplet concentration from 0 to 5 wt % but then increased less steeply at higher values. The *a* and *b* values became increasingly negative as the droplet concentration was increased up to about 0.5-1 wt % oil (depending on the droplet size) but became less negative above this value. This suggests that the emulsion initially became more blue and more green with increasing droplet concentration up to a certain level and then became less blue and less green with a further increase.

The most significant change in emulsion color (relative to the color of emulsions in the absence of droplets) occurs between 0 and 5 wt % oil (Figure 8f). At higher droplet concentrations the change in color is much less appreciable. The hue angle and chroma increase with increasing droplet concentration up to a certain value and then decrease (Figure 8d,e).

The maximum in the intensity of the emulsion color at a particular droplet concentration (as indicated by the maximum in the chroma) is most likely caused by changes in the relative contribution of absorption and scattering to the overall emulsion appearance as droplet concentration varies (Figure 9). In the absence of droplets, the appearance is determined by the absorbance of light which has traveled through the solution and been reflected from the white plate. When droplets

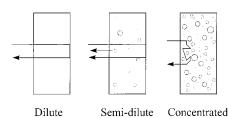


Figure 9. Contribution of scattering and absorption to emulsion appearance dependence on droplet concentration.

are present some of the light is scattered back to the detector, which decreases the effective path length that the light travels through the dye solution and therefore reduces the overall degree of absorption. At high droplet concentrations, the scattering is so intense that the light wave is unable to penetrate very far into the emulsion before being reflected back, and so even less light is absorbed. Previous studies have observed a maximum in the *a* and *b* values when the pigment concentration of a solution is increased (Eagerman, 1972), which is equivalent to increasing the effective path length of light in the dye solution in our system.

In general, the influence of droplet concentration on the lightness and color of emulsions containing different droplet sizes follows a similar trend (Figure 8). Nevertheless, the precise shapes of the curves do depend on droplet size. For example, the position and depth of the minimum in the *a* and *b* values vary with mean droplet diameter. Differences in emulsion appearance due to differences in droplet concentration could be distinguished by our untrained panel (Table 2). Panelists found a trend toward decreasing "blueness" and increasing "lightness" as the droplet concentration increased. A number of the panelists ranked the "lightness" of the 10 and 15 wt % emulsions in the opposite order than was expected. This finding would support the instrumental color measurement which indicated that the *L* value, which is related to lightness, does not change appreciably between 10 and 20 wt % (Figure 8a). Our results clearly indicate that droplet concentration has a major impact on emulsion appearance.

Linear regression analysis showed a strong correlation between *L* value and panelist score of "lightness" (r = -0.874), between *b* value and droplet concentration (r = 0.910), and between *b* value and panelist score of "blueness" (r = 0.914) and a reasonable correlation between *L* value and droplet concentration (r = -0.73).

CONCLUSIONS

This study has shown that droplet characteristics have a pronounced influence on emulsion appearance. The "lightness" of an emulsion is correlated to the scattering efficiency of the droplets, which is related to both their size and concentration. Our results suggest that there is a maximum in the scattering efficiency at a droplet diameter somewhere less than a micrometer. The maximum in emulsion turbidity (which occurred at $d_{32} \sim 2 \ \mu$ m) did not correlate well with emulsion lightness (as measured instrumentally or assessed by a sensory panel) because it relies on measurements of the light which has been transmitted through an emulsion, rather than that which has been reflected.

As the droplet size increased, the scattering efficiency of the droplets decreased, which caused a reduction in emulsion lightness and an enhancement of emulsion color. On the other hand, as the droplet concentration increased, there was an increase in emulsion lightness and a reduction in emulsion color. The effects of droplet concentration were most pronounced in the range 0-5wt % oil. Our results have important implications for the formulation of food emulsions. To obtain an emulsion with a specific appearance it is necessary to carefully control both the size and concentration of the emulsion droplets, as well as the amount of dye used. In addition, the development of an objective method of characterizing emulsion color will be extremely valuable for fundamental studies of the factors which influence emulsion color and for the assurance of product quality during manufacturing processes.

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