

## Retention of Emulsified Flavor in a Single Droplet during Drying

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**The prevention of the loss of flavor compounds during spray drying is one of the most important engineering techniques to obtain high quality powdery food products. In the present study, a droplet drying technique was used to evaluate the mechanism of the loss of hydrophobic liquid flavors from a droplet containing flavor emulsions in an aqueous mixture solution of gum arabic and maltodextrin. The effects of type of flavor, air temperature, the initial concentration of carrier solids and the stability of the emulsion on the retention of different kinds of flavors were investigated. The retention of flavor was markedly dependent on the type of flavor and the stability of emulsion, and retention could be correlated well with emulsion stability. A simple mathematical model of the loss of emulsified flavor during drying was proposed, based on a breakdown hypothesis of the emulsion droplet.**

Keywords: flavor retention, spray drying, emulsion, diffusion

Spray drying is the one of the most cost effective techniques for the production of powdery foods from liquid food materials. To make high quality products, prevention of the loss of volatile ingredients is of the utmost importance during spray drying. The most common and widely used technique to encapsulate hydrophobic flavors is to emulsify the flavor and then spray-dry the suspension liquid of the emulsion. Many investigations (Zakarian & King, 1982; Brenner, 1983; Rosenberg *et al.*, 1990; Shue & Rosenberg, 1995; Rosenberg & Shue, 1996; Bhandari *et al.*, 1992; Liu *et al.*, 1999) have been made to elucidate the mutual relation between the process variables of a spray drying system and the retention of the emulsion flavor during the spray drying. Coumans *et al.* (1994), King (1995), and Ré (1996) have given extensive overviews on the theoretical and practical aspects of flavor retention, and the process variables and mechanisms governing the losses of flavor compounds in spray drying. Two symposium books have also been published concerning flavor encapsulation and release of the encapsulated flavors (Risch & Reinneccius, 1988 and 1995). However, it is quite difficult to evaluate independently the influence of certain process variables of a spray drying system on flavor retention. To investigate the drying behavior of droplets inside a spray dryer, a single droplet drying method has been used to study the mechanism of flavor retention during drying. Furuta *et al.* (1984) investigated the retention of a trace amount of ethanol, suggesting that the selective diffusion theory (Thijssen & Rulkens, 1968) could be successfully applied to estimate the amount of ethanol retained during drying. Verderber and King (1992) and Sunkel and King (1993) carried out simultaneous measurements of flavor loss and the morphological change of the droplet during drying, reporting that substantial loss of flavor takes place due to rapid cycles of expansion, bursting and cratering of the droplet during drying. Sayed *et al.* (1996), Hassan *et al.* (1996a; b), and Walton and Mumford

(1999) studied the crust and the inside structure of the dried droplet using a scanning electron microscope, and related the morphological change with the loss of ethanol during drying of a single droplet. Liu *et al.* (2000) investigated the retention of *l*-menthol included in cyclodextrin during drying of a single droplet. They observed that the crust formation of carbohydrate is important for the retention of the included *l*-menthol. However, less research has been done on the mechanism of the retention of emulsified flavors. Imagi *et al.* (1992) and Minemoto *et al.* (1997) applied a single droplet drying method to evaluate the oxidative mechanism of spray-dried unsaturated fatty acid during storage. The droplet to be dried was placed at a constant temperature and humidity and influence of the combination of wall material and emulsifier on the oxidation of the emulsified fatty acid in the droplet was evaluated.

In the present study, a droplet drying technique was applied to determine the loss of flavors during drying of a droplet containing flavor emulsion. The effects of the type of flavor, air temperature, and the concentration of maltodextrin on variations in flavor retention were investigated. We have proposed a simple mathematical model to calculate the final retention of flavor, based on the first order emulsion rupture model and the selective diffusion of the dissolved flavor.

### Materials and Methods

**Materials** *d*-Limonene, ethyl butyrate, ethyl propionate and gum arabic (GA) were purchased from Nacalai Tesque (Kyoto), and ethyl caproate from Tokyo Kasei Kogyo Co., Ltd. (Tokyo). Maltodextrin (MD; dextrose equivalence is 18) was a gift from Nippon Starch Chemicals Co., Ltd. (Osaka). The organic chemicals used in the analyses were of analytical grade.

**Preparation of emulsion solution of flavor** *d*-Limonene, ethyl caproate, ethyl butyrate, and ethyl propionate were used as model flavors. Carrier solution was prepared by blending and rehydrating maltodextrin with gum arabic in different proportions in warm distilled water, followed by cooling to room tem-

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perature in chilled water. The total solid concentration of the carrier solution ranged from 10 to 30% (w/w). Seven grams of the carrier solution was put into a test tube (15×120) and blended with 0.18 g of a flavor. The mixture was homogenized for 3 min at power control 5 by a Polytron homogenizer (PT-10, Kinematica GA, Littau, Switzerland) consisting of a stator and rotor with a blade. During homogenization, the test tube was cooled with ice.

**Drying of a single droplet containing emulsion** The experimental apparatus for drying of a single droplet containing emulsion was the same as that used previously (Furuta *et al.*, 1984). In a vertical wind tunnel, air temperature, velocity, and humidity were controlled by an assembly of a packed bed humidifier and a regulated electric heater. At the outlet of the wind tunnel, an upstream airflow was maintained at a uniform temperature and velocity by means of a contraction nozzle. Precisely 5  $\mu\text{l}$  of the emulsion solution was measured with a microsyringe, and suspended at the tip of a glass filament in the form of a small pendant. The glass filament was hung in the upstream hot air. After drying for the designated time, the droplet was removed from the hot air and put into a mini-vial (0.3 ml with a small stirrer bar) containing 200  $\mu\text{l}$  of hexane and 50  $\mu\text{l}$  of distilled water to extract the remaining flavor into hexane. The time required for the extraction was 90 min for *d*-limonene and 30 min for the esters, respectively. The flavor content in hexane was determined by a gas chromatograph (GC-14B, Shimadzu, Kyoto) equipped with a flame ionization detector. A glass column (2.5 m×2 mm I.D.) packed with PEG 20M on 80/100 mesh chromosorb W (AW-DMCS) (Shimadzu) was used. The column temperature was 110°C and the flow rate of the nitrogen carrier gas was 50 ml/min. The retention of flavor was defined as the ratio of flavor content in the droplet before and after drying. A thermocouple (alumel-chromel, 100  $\mu\text{m}^{\phi}$ ) was used to measure the temperature history of the droplet during drying. The electromotive force from the thermocouple was recorded with a pen recorder (L-4120E, YOKOGAWA, Tokyo) to determine the surface evaporation period of drying.

**Measurement of apparent diffusivity of flavor dissolved in the solution** To measure the apparent diffusivity of flavor in the carrier solution, measurements were made for single droplet drying experiments using a droplet containing no emulsion (i.e., a droplet composed of the saturated solution of esters with dissolved solid). The diffusivity could be determined by the surface evaporation interval and the final retention of flavor. Assuming that all the loss of flavor takes place during the surface evaporation period, the apparent diffusivity of the dissolved flavor in the carrier solution,  $D_{\text{app}}$ , can be estimated by the following equation (Crank, 1975):

$$\frac{C_A}{C_{A0}} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left\{-D_{\text{app}}\left(\frac{\pi n}{R}\right)^2 t_c\right\}, \quad (1)$$

where  $C_A/C_{A0}$  is the flavor retention,  $t_c$  the surface evaporation interval and  $R$  is the radius of the droplet, which is assumed to be constant during  $t_c$ . Since the interval of the surface evaporation period  $t_c$  could be easily obtained by measurement of the droplet temperature, the value of  $D_{\text{app}}$  can be obtained from Eq. (1).

**Measurement of emulsion stability** The stability of the emulsion was measured, basically following the turbidimetric technique proposed by Pearce and Kinsella (1978). The emulsion

solution prepared as described above was diluted with 0.1% (w/v) sodium dodecyl sulfate (SDS) solution and absorbance at 500 nm was measured by a spectrophotometer (UV-1200, Shimadzu). The emulsion solution was put into a test tube, which was incubated in a water bath at 25°C. At designated intervals, 100–600  $\mu\text{l}$  of the solution was taken from the bottom of the test tube and diluted with 0.1% SDS solution to 1/13 to 6/13 to adjust the initial absorbance of emulsion to 0.1 to 1.5. Stability of the emulsion was defined as the decreasing rate of absorbance in the initial period.

## Results and Discussion

**Time course of the flavor retention and temperature history of droplet during drying** Figure 1 shows the time-course of the retention of *d*-limonene, ethyl caproate, ethyl butyrate, and ethyl propionate during the drying of a single emulsion droplet in hot air at 70°C. The composition of the carrier solution was GA 10% and MD 10%. It was found that the retention of ethyl propionate and ethyl butyrate decreased rapidly during the initial period of drying for 2 to 3 min, and then remained constant after 3 min of drying, while that of *d*-limonene and ethyl caproate gradually decreased in the initial period followed by a constant retention. Of the flavors used in this study, *d*-limonene had the highest final retention (after 20 min drying) followed by ethyl caproate, ethyl butyrate, and ethyl propionate, indicating that the final retention decreased in the order of the polarity of the flavor.

In Fig. 1 the temperature history of the droplet containing ethyl butyrate emulsion is also illustrated by a dotted line. The temperature increased at the onset of drying, followed by a constant temperature period, after which it increased markedly. Considering the retention time course coupled with the droplet temperature history, we found that over 90% of the total loss of the flavor took place during this period. The retention time-courses of the emulsified flavors were qualitatively quite similar to those reported by Furuta *et al.* (1984) for ethanol in maltodextrin solution and Liu *et al.* (2000) for *l*-menthol included in  $\beta$ -cyclodextrin.

**Effect of air temperature on retention of flavor** Figure 2 shows the effect of increasing air temperature on the final reten-

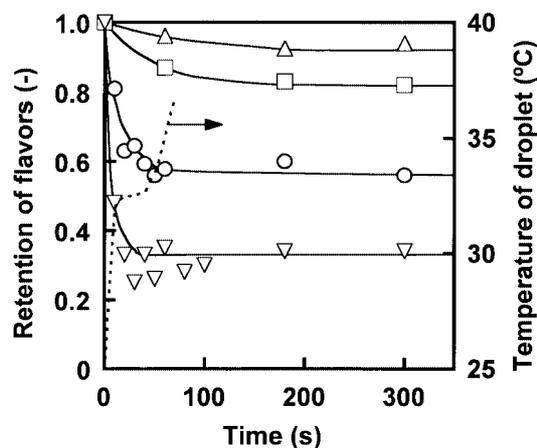


Fig. 1. Retention time course of flavors and temperature history of the droplet during single droplet drying.  $\Delta$ , *d*-limonene;  $\square$ , ethyl caproate;  $\circ$ , ethyl butyrate;  $\nabla$ , ethyl propionate. ----, Droplet temperature. Air temperature = 70°C. GA = 10% and MD = 10%.

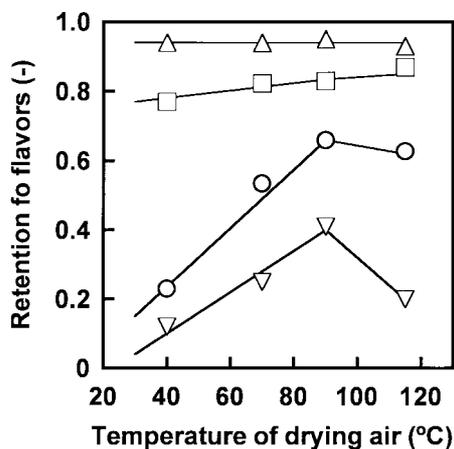


Fig. 2. Effect of air temperature on the retention of flavors. Symbols are the same as in Fig. 1. GA=10% and MD=10%.

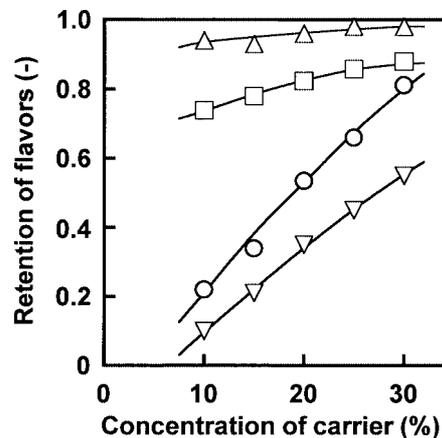


Fig. 3. Effect of initial solid concentration on the retention of flavors. Symbols are same as in Fig. 1. Air temperature=70°C. GA=10%. MD=0–20%.

tion of the flavor. The retention of *d*-limonene was independent of air temperature between 40 and 100°C. In contrast, the retention of ethyl caproate slightly increased as the air temperature increased from 40 to 100°C, while marked increases in the retention of ethyl butyrate and propionate retention were observed. The emulsions of ethyl butyrate and ethyl propionate were so unstable (Liu *et al.*, 1999) that the emulsion was assumed to break inside the droplet during drying, and the emerging flavor to diffuse through the droplet for subsequent evaporation from the surface. However, when the air temperature was high, a crust covered the surface of the droplet so quickly that the flavor could not evaporate easily from the surface (Thijssen & Rulkens, 1968; Furuta *et al.*, 1984; Liu *et al.*, 2000). This resulted in a steep increase of the retention of ethyl butyrate and ethyl propionate with an increase in air temperature. For *d*-limonene or ethyl caproate, the retention was high and independent of the temperature of hot air, due both to the stable emulsion and the low solubility.

At the air temperature of 115°C, the droplet experienced a morphological change of cyclic inflation and bursting. However, the retentions of *d*-limonene and ethyl caproate were not influenced by the morphological changes. The retention of ethyl butyrate and ethyl propionate, however, decreased abruptly at the air temperature of 115°C. This might have resulted from both breakdown of the flavor emulsion at inflation or bursting of the droplet, and the increasing droplet temperature above or near the boiling point of these flavors.

**Effect of initial carrier solid concentration** The effect of the initial carrier solid concentration on the flavor retention is shown in Fig. 3. The x-axis represents the total initial solid concentration of gum arabic and maltodextrin in the emulsion solution. *d*-Limonene showed the highest retention of more than 95% and was independent of the initial carrier solid concentration. The retention of ethyl caproate slightly depended on the initial carrier solid concentration. For ethyl butyrate and ethyl propionate, on the other hand, the retention was markedly affected by the solid concentration. Below 25% solids, particularly, the retention increased steeply with the increase in concentration. At high solid concentration the formation of crust on the surface of the droplet was so rapid that the ethyl butyrate emerging from the

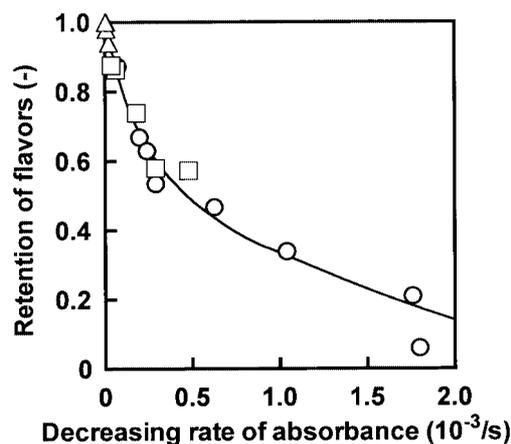


Fig. 4. Retention of flavors as the function of emulsion stability. Symbols are the same as in Fig. 1. GA=10%. MD=10%. Air temperature=70°C.

ruptured emulsion was trapped. This implies that the selective diffusion phenomenon proposed by Thijssen and Rulkens (1968) took place for ethyl butyrate and ethyl propionate (Furuta *et al.*, 1984; Liu *et al.*, 2000).

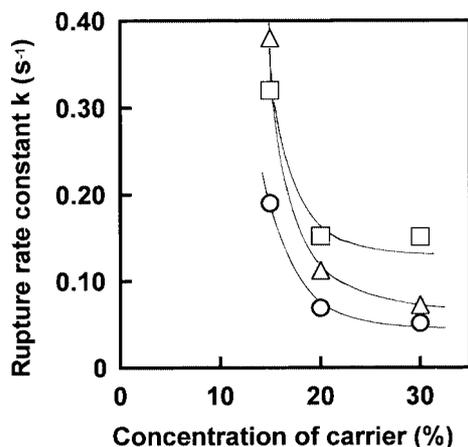
**Correlation of flavor retention to the stability of emulsion** In Fig. 4 the final retentions of flavors at 70°C under various compositions of the carrier solution are plotted against the decreasing rate of emulsion absorbance at the initial period. For each flavor, the final retention correlated well with the decreasing rate of emulsion absorbance. The correlation indirectly demonstrated that the unstable emulsion was broken inside the droplet, resulting in appreciable loss of flavor during drying. Since the absorbance of ethyl propionate emulsion was too low to determine the reliable time-course of emulsion absorbance, the final retention of ethyl propionate could not be correlated by this method.

**Emulsion rupture model for evaluation of the retention of ethyl butyrate during drying of a single droplet** To estimate the final retention of flavor of an unstable emulsion such as ethyl butyrate or ethyl propionate, a simple mathematical model was developed on the basis of the emulsion rupture assumption. In

**Table 1.** Effective diffusivity of ethyl butyrate and the rupture rate constant for calculations.

Air temperature (°C)	Wet-bulb temperature (°C)	Maltodextrin conc. (% w/w)	Surface evaporation period $t_c$ (s)	Apparent diffusivity $D_{app} \times 10^9$ (m <sup>2</sup> /s)	Rupture rate constant $k$ (s <sup>-1</sup> )
40	24	5	106	1.97	0.19
		10	83	1.62	0.069
		20	46	0.639	0.051
70	31.5	5	37	1.99	0.38
		10	27	1.78	0.11
		20	16	0.735	0.072
90	36.2	5	23	2.05	0.32
		10	17	1.31	0.15
		20	11	0.714	0.15

The initial concentration of gum arabic was 10% (w/w).



**Fig. 5.** Rupture rate constant as functions of the initial concentration of carrier solid and air temperature. ○, Air temperature=40°C; △, 70°C; □, 90°C.

this study, ethyl butyrate was selected as an unstable flavor emulsion. For simplicity, the following assumptions were made as to the loss of ethyl butyrate during the drying of a single droplet.

i) From Fig. 1 it is found that the major loss of ethyl butyrate took place during the surface evaporation period. Therefore, one can assume that the flavor loss was complete at the end of the surface evaporation period.

ii) The emulsions were so unstable that the emulsion particles might have ruptured inside the droplet during drying. A first order rate was assumed for the rupture of the emulsion.

iii) The radius of the droplet  $R$  is assumed to be constant during the surface evaporation period  $t_c$ .

On the basis of the above assumptions, the change of ethyl butyrate dissolved in the droplet can be written as Eq. (2):

$$\frac{\partial(1-\phi)C_A}{\partial t} = (1-\phi)D_{app} \left( \frac{\partial^2 C_A}{\partial r^2} + \frac{2}{r} \frac{\partial C_A}{\partial r} \right) + v_{rupt}, \quad (2)$$

where  $C_A$  is the flavor concentration dissolved in the carrier solution,  $D_{app}$  the apparent diffusivity of flavor,  $v_{rupt}$  the rate of the rupture of the emulsified flavor,  $\phi$  the volume fraction of flavor emulsion, and  $r$  the radial coordinate. Following the second assumption, the rate of emulsion rupture  $v_{rupt}$  can be described as Eq. (3):

$$v_{rupt} = -\tilde{C}_E \frac{d\phi}{dt} = k\tilde{C}_E\phi = k\tilde{C}_E\phi_0 \exp(-kt), \quad (3)$$

where  $k$ ,  $\phi_0$  and  $\tilde{C}_E$  are the rupture rate constant, the initial vol-

ume fraction of flavor emulsion, and molar concentration of the pure flavor, respectively. The initial and boundary conditions of Eq. (2) are as follows:

$$C_A = C_{A0}, \text{ at } t=0; C_A = 0, \text{ at } r=R; \partial C_A / \partial r = 0 \text{ at } r=0, \quad (4)$$

where  $C_{A0}$  is the initial concentration of ethyl butyrate in the dissolved state.

The final retention of ethyl butyrate during drying of a single droplet was calculated according to the following procedure: Using the experimental values of  $t_c$  and the final retention of dissolved ethyl butyrate, the apparent diffusivity of ethyl butyrate  $D_{app}$  was calculated by Eq. (1), and is listed in Table 1 for different temperatures and maltodextrin concentrations. The finite-difference representation of Eq. (2) was obtained by Crank-Nicolson's implicit approximation (Smith, 1965) to calculate the concentration profile of the dissolved flavor inside the droplet during drying. The mesh sizes in  $r$ -direction and time were 1/20 and 1/1000, respectively. The concentration profile of the dissolved flavor in the droplet at time  $t_c$  could then be calculated. The sum of the total amount of the dissolved and emulsified flavor in the droplet at  $t_c$  was divided by the initial amount of flavor to calculate the retention of flavor, as:

$$Retention = \frac{3(1-\phi) \int_0^R r^2 C_A dr + R^3 \phi \tilde{C}_E}{R^3 [(1-\phi_0)C_{A0} + \phi_0 \tilde{C}_E]}. \quad (5)$$

The rupture rate constant  $k$  in Eq. (3) was evaluated so as to coincide with experimental final retention of ethyl butyrate with the calculated value of Eq. (5). For the various initial compositions of the carrier solution and the drying conditions, we found the optimal rupture rate constants of ethyl butyrate, which are listed in Table 1 for different temperatures and solid concentrations in carrier solution. Figure 5 shows the rupture rate constant  $k$  against the concentration of solid in the carrier solution. The value of  $k$  sharply decreased with increase of the initial concentration of carrier solid between 15% to 20%, indicating that the emulsion became more stable with increase in the solid content of the solution (Liu *et al.*, 1999). The rupture rate constant was also influenced by temperature, increasing with an increase in air temperature as shown in Table 1. By Arrhenius plot of  $k$  against the reciprocal of the wet-bulb temperature of the drying air, the activation energies of  $k$  were obtained as 36, 50, and 64 kJ/mol at the initial carrier solid concentrations of 15, 20, and 30 % (w/w), respectively.

## Conclusions

A single droplet drying technique was used to investigate the

mechanism of the loss of hydrophobic liquid flavors from a droplet containing emulsions of flavor by an aqueous solution composed of gum arabic and maltodextrin. The retention of flavor was dependent on the type of flavor and the concentration of GA and MD. The stability of flavor emulsion droplet was also a key factor in the retention of flavor. The retention of flavors correlated well with the stability of the emulsion droplet. A simple mathematical model of the loss of emulsified flavor during drying was proposed, based on the rupture of the emulsion droplet during drying. The rupture rate constant used for the calculations decreased with an increase in the initial concentration of carrier solid, indicating that the emulsion became stable at high concentrations of solid.

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