Process of Forming Seamless Capsules by Concentric Nozzle System†

Toshiyuki Suzuki, Hideki Sunohara
and Ryosei Kamaguchi
Morishita Jintan Co., Ltd.*

Abstract

Adaptation of the natural phenomenon of drop formation has resulted in the development of forming seamless capsules.

The encapsulation behavior of gelatine in coolant fluid has been studied using a concentric nozzle. The major assumption was that capsules were formed through a wave-like instability in the concentric streams of extruded core, shell, and coolant fluids.

Predictions of flow rates and fluid properties were compared with experimental results and were shown to be in reasonably good agreement. It was found that for a given set of flow rates and fluid properties optimum conditions could result in forming uniform size capsules.

1. Introduction

One of the advantages of a capsule is to apparently treat formless liquid as a solid form. A soft capsule which is formed by concentric nozzle system based on an orifice method is called a seamless mini-capsule, and has been widely used for a variety of industries such as food, pharmacy, and so on. This capsule shows excellent powder characteristics of fluidity and packing behavior since it is formed to have spherical shape. However, the influence of physical property on capsule formation process has been scarcely investigated. This paper presents experimental findings on this influence with the acid of a fundamental model concept.

2. Experimental equipment

The experimental equipment used in this work is schematically illustrated in Fig. 1.

Shell liquid and core liquid stored in each tank ① and ② respectively were introduced quantitatively into a concentric nozzle ⑤ by gear pumps ③ and ④ and discharged out of the tip of the nozzle so that a jet flow ⑧ could be formed in a guide tube ⑦. When the flow of coolant oil filled in this guide tube was laminar, the stable elongation of the jet flow would be permitted to make sequential formation of resultant capsules. In this process, moreover, a vibration ring ⑪ was equipped to form uniform size capsules.

On the other hand, the coolant oil to be used for forming capsules by cooling was introduced into the guide tube ⑦ mounted at the center of a cooling cylinder ⑥, after being cooled in a heat exchanger ⑫. This liquid accompanied by formed capsules was transported through a cooling tube ⑭, before being made to be separated from the capsules by separator ⑬ placed at the outlet of the tube. After this separation, the liquid was stored in a coolant oil tank ⑮ and returned to the heat exchanger ⑫ by gear pump ⑯.

3. Experimental procedure and results

In this work, the influence of each liquid flow rate and shell liquid viscosity on capsule formation process was experimentally investigated. For simplification, the core liquid, the shell liquid, and the coolant oil will be called...
The formation of capsules is schematically outlined in Fig. 2. The jet length $L$ is considered to be a distance where the jet flow of liquids 1 and 2 is capable of maintaining its stability. In this case, the diameter of this jet flow at the point which is $L/2$ distant from the tip of the nozzle may be reasonably regarded as a standard dimension of the flow; however it should be noticed that the diameter at an arbitrary point is dependent upon the vertical distance in the flow.

Relationship between jet diameter $\bar{d}_j$ and capsule diameter $d_c$

The influence of jet diameter $\bar{d}_j$ on capsule diameter $d_c$ was experimentally studied under the conditions listed in Table 1. As shown in Fig. 3, plots of the results hardly deviated from the single straight line which demonstrated that $d_c$ would be twice as large as $\bar{d}_j$. It is found that this result might be in good agreement with the solution derived from the Rayleigh theory for instability of liquid jets.  

1st liquid flow (core liquid flow)

2nd liquid flow (shell liquid flow)

3rd liquid flow (coolant liquid flow)

Fig. 2 Schematic of capsulation

Fig. 1 Schematic diagram of apparatus
Table 1 Experimental conditions

| Condition | 1st liquid flow rate $q_1 \, [\text{g/s}]$ | 2nd liquid flow rate $q_2 \, [\text{g/s}]$ | 3rd liquid flow rate $q_3 \, [\text{g/s}]$ |
|-----------|-----------------|-----------------|-----------------|
| 1         | 0.653           | 0.531           | 16.6            |
| 2         | 0.557           | 0.531           | 26.3            |
| 3         | 0.477           | 0.531           | 36.0            |
| 4         | 0.374           | 0.531           | 45.7            |
| 5         | 0.281           | 0.531           | 55.4            |
| 6         | 0.194           | 0.531           |                 |

Fig. 3 Correlation of capsule diameter $d_c$ and jet diameter $d_j$

**Capsule formation**

Consider a typical capsule model during its successive formation as illustrated in Fig. 4. Provided that the distance between the successively formed two capsules (pitch $P$) is assumed to be the length of the cylindrical jet flow required to make one capsule, the following equation is given:

$$\pi \left( \frac{d_j}{2} \right)^2 P = \frac{4}{3} \pi \left( \frac{d_c}{2} \right)^3$$  \hspace{1cm} (1)

As shown in Fig. 5, the experimental results based on the conditions presented in Table 1 seem to be little apart from the linear relation of Eq. (1). It follows that Eq. (1) could reasonably relate the jet diameter $d_j$ and the capsule diameter $d_c$, which was also confirmed by Dabora\(^2\). From Eq. (1), the capsule diameter $d_c$ can be rewritten as a function of pitch $P$ as well as jet diameter $d_j$ as follows.

$$d_c = \left(\frac{3}{2} \frac{d_j^2}{P}\right)^{\frac{1}{3}}$$  \hspace{1cm} (2)

This equation states that capsule diameter $d_c$ can vary with jet diameter $d_j$ alone, if pitch $P$ is held constant. Pitch $P$ may be held constant, if vibration is applied to the jet flow. By the way, under the conditions listed in Table 1 the encapsulation processes observed is schematically illustrated in Fig. 6; the capsule diameter decreased with increasing the value of $q_3$. On the other hand, it was found that the jet diameter $d_j$ increased with increasing the values of $q_1$ and/or $q_2$.

**Relationship between flow rate ratio $(q_1 + q_2)/q_3$ and jet diameter $d_j$**

From the facts mentioned above, it can be known that liquids 1 and 2 might be positive factors for rising the jet diameter $d_j$ and liquid 3 might be a negative one. Figure 7 shows the
dependence of the flow rate ratio \( (q_1 + q_2)/q_3 \) upon the jet diameter \( d_j \).

When it is assumed that all the liquids are Newtonian and flow at the same rate, the following equation can be given:

\[
\frac{q_1}{\gamma_1} + \frac{q_2}{\gamma_2} = \frac{\pi}{4} d_j^2 V_{3 \text{ max}}
\]  
(3)

where \( \gamma_i \) is the specific weight in g/cm\(^3\) for the \( i \)-th liquid. The maximum flow rate of liquid 3 is expressed as

\[
V_{3 \text{ max}} = \frac{8 q_3}{\pi \gamma_3 d_3^2}
\]  
(4)

The experimental condition employed in this work was that \( \gamma_1 = \gamma_2 = 1 \times 10^{-3} \text{ g/mm}^3 \), \( \gamma_3 = 0.94 \times 10^{-3} \text{ g/mm}^3 \), and \( d_3 = 20 \text{ mm} \). Thus Eqs. (3) and (4) can be rewritten as the following relation:

\[
\frac{1}{d_j^2} = \frac{1}{48} \left( \frac{q_1}{q_3} + \frac{q_2}{q_3} \right)
\]  
(5)

It is obvious from Fig. 7 that the plots obtained experimentally in this work is in good agreement with the solution of Eq. (5) indicated as the dotted curve. This suggests that the assumption in the above was consistent with the result of this work.

Relationship between flow rate ratio \( (q_1 + q_2)/q_3 \) and capsule diameter \( d_c \)

To find the dependence of the flow rate ratio \( (q_1 + q_2)/q_3 \) upon the capsule diameter \( d_c \), another encapsulation experiment was carried out under the condition of the temperatures of 22, 80, and 10°C for liquids 1, 2, and 3 respectively and the concentration of 25% for liquid 2. The experimental condition and the result are indicated in Table 2 with the
Table 2 Flow rate ratio \((q_1 + q_2)/q_3\) and capsule diameter \(d_e\)

| 1st liquid \(q_1\) [g/s] | 2nd liquid \(q_2\) [g/s] | 3rd liquid \(q_3\) [g/s] | \((q_1 + q_2)/q_3\) [-] | Minimum | Mean | Maximum |
|-------------------------|-------------------------|-------------------------|----------------------|---------|------|--------|
| 0.1473                  | 0.3057                  | 38.107                  | 0.01189              | 2.70    | 3.80 | 4.30   |
| 0.3180                  | 0.3057                  | 30.203                  | 0.02065              | 2.10    | 3.90 | 5.95   |
| 0.4318                  | 0.4151                  | 30.203                  | 0.02804              | 2.10    | 4.35 | 5.25   |
| 0.3180                  | 0.3057                  | 14.395                  | 0.04332              | 3.95    | 5.49 | 7.00   |
| 0.4318                  | 0.4151                  | 14.395                  | 0.05883              | 5.75    | 6.71 | 8.10   |
| 0.5171                  | 0.4972                  | 14.395                  | 0.07046              | 3.25    | 7.01 | 9.85   |
| 0.5740                  | 0.5519                  | 14.395                  | 0.07821              | 3.00    | 7.35 | 9.50   |

schematic illustration of Fig. 8. It is found from this figure that the flow rate ratio \((q_1 + q_2)/q_3\) was approximately proportional to the capsule diameter \(d_e\) with a region of \(0.02 < (q_1 + q_2)/q_3 < 0.06\). This implies that there could be an optimum flow rate ratio for formation of the capsule with a uniform diameter.

**Influence of flow rate ratio \(q_1/q_2\) on mean capsule diameter \(d_e\)**

As shown in Fig. 9, the mean capsule diameter \(d_e\) was almost independent of the ratio of the flow rate of liquid 1 to that of liquid 2, \(q_1/q_2\).

**Influence of liquid 2 concentration on mean capsule diameter**

The mean capsule diameters are plotted against the concentration of liquid 2 as a function of \((q_1 + q_2)/q_3\) in Fig. 10. It is found that the diameter decreased with increasing the concentration and this tendency became more remarkable as \((q_1 + q_2)/q_3\) increased. This is probably because the spherical formation of the capsules was enhanced both by the increase in the surface tension of liquid 2 due to the rise of its concentration and by increase in the flow rate of liquid 3 which resulted in reduced jet diameter.

The physical factors investigated in this work were the flow rate of the liquids and the viscosity of liquid 2 and they might be also dependent on a more fundamental factor, i.e. temperature. Therefore further study on the influence of temperature should be required to control the capsule formation process more precisely, although only the restricted range of the temperature effect was investigated in this work.

4. **Conclusion**

It has been little known how the two kinds of liquid are discharged out of the concentric nozzle into another liquid in a jet flow. In this work, we investigated the influence of these three liquids on the diameter of capsule formed in the solidification process in order to elucidate such a complicated fluid dynamic behavior. The results of experiments and observations are summarized below.

1) The capsule diameter \(d_e\) is about twice as large as the jet diameter \(d_j\).
2) The capsule diameter \(d_e\) is dependent upon the pitch \(P\) as well as the jet diameter \(d_j\), and thus \(d_e\) may be held constant if vibration is applied on the flow to fix the value of \(P\).
3) There is an optimum flow rate ratio \((q_1 + q_2)/q_3\) which yields the capsules with a uniform diameter.
4) The capsule diameter \(d_e\) decreases with an increase in the viscosity of the shell liquid.
5) The diameter of stabilized capsules may be determined when the velocity of the jet flow coincides with that of the coolant oil.

Based on this work, we intend to investigate the influence of other physical factors such as temperature, specific weight, and so on.

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Nomenclature

- $d_c$: capsule diameter [mm]
- $\bar{d}_c$: mean capsule diameter [mm]
- $d_j$: jet diameter [mm]
- $d_3$: inner diameter of guide tube [mm]
- $L$: jet length [mm]
- $P$: pitch [mm]
- $q_1$: flow rate of core liquid [g/s]
- $q_2$: flow rate of shell liquid [g/s]

- $q_3$: flow rate of coolant oil [g/s]
- $V_{3\text{max}}$: maximum velocity of coolant oil [mm/s]
- $\gamma_1$: specific weight of core liquid [g/cm$^3$]
- $\gamma_2$: specific weight of shell liquid [g/cm$^3$]
- $\gamma_3$: specific weight of coolant oil [g/cm$^3$]

References

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