Microgranulation and Encapsulation of Pulverized Pharmaceutical Powders with Ethyl Cellulose by the Wurster Process

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A pulverized phencacetin powder with a mass median diameter of 11 μm was slightly granulated and subsequently coated to produce fine microcapsules by the Wurster process. As a membrane material, a 1:1 mixture of ethyl cellulose (EC), cholesterol (CH) and talc was used in the granulation, and a 1:1:0.06 mixture of EC, CH and stearyltrimethylammonium chloride in the coating. The produced microcapsules had a mass median diameter of 31 μm at 60% coating level and exhibited a practically useful sustained release at 40% or more coating.

In the granulation process, the membrane material solution was sprayed at a high rate of 14.2 ml/min upon 250 g of phencacetin powder. The mass median diameter was increased to 19 μm at a binder level of 3% (EC plus CH). The granules were then dried to reduce the particle size to 14 μm. This microgranulation and drying were effective for steady fluidization of the fine powder and to avoid excessive agglomeration. Coating was performed at a lower spray rate of 11.3 ml/min under a stronger agitation to avoid excessive agglomeration. During the coating process, however, an increase in the mass median diameter by 17 μm and the broadening of size distribution were unavoidable. The smallest size of particles which could be coated with no subsequent agglomeration was estimated to be 20 μm. The agglomeration of the particles smaller than 20 μm which occupied 78% of the raw powder accounted for the particle growth and the broadening of size distribution.

Keywords granulation; microcapsule; coating; ethyl cellulose; cholesterol; stearyltrimethylammonium chloride; Wurster process; powder; phencacetin; dissolution

The Wurster process is characterized by fine particle coating, and is a favorable technique for microencapsulation. However, there remain some problems to solve before it can be used practically.

In a previous paper, calcium carbonate of 32—44 μm was layered with phencacetin and subsequently microencapsulated with a mixture of ethyl cellulose, cholesterol and stearyltrimethylammonium chloride (2:1:0.03). The produced microcapsules had a mass median diameter of 56 μm. In this study, the coating of finer pharmaceutical powders of the order of 10 μm was attempted. The previous calcium carbonate had a high density of 2.93 g/cm³. In the Wurster process, the particles blown up from partition have to fall down gravimetrically, therefore, heavier particles are more steadily fluidized. However, pharmaceutical powders very often have low particle densities, in addition, they are sometimes electrostatically charged, leading to retardation in particle recycling and to production of poorly coated microcapsules. Although the Wurster method can process fine powder, it appeared that coating as fine a pharmaceutical powder as 10 μm as discrete particles would be difficult.

Ordinary top-sprayed granulation in a fluidized bed produces agglomerates with low apparent density and irregular, rough surface. Since such agglomerates are not appropriate for subsequent coating, an additional consolidation and surface-smoothing by means of mechanical agitation are usually required. As is well known, the particle motion in the Wurster process is very rapid, and particles collide with the inner surface of the partition, leading to the disintegration of large, weakly flocculated agglomerates. Thus, this process can be expected to produce very fine, dense granules. If original particles were not coated as discrete particles and would unavoidably be agglomerated because of their fineness and low density, such dense agglomerates should be efficiently coated to produce fine microcapsules. Hence, the process in this study was designed for the fine powder to be first granulated and subsequently coated. Throughout both processes, materials known to be active as a permentation barrier were sprayed. The water soluble polymer used in usual fluidized granulation was not used.

Experimental

Materials As a model drug, phencacetin (Kawasaki Kagaku Kogyo Co., Ltd.) was used. The course phencacetin powder purchased was pulverized with a Sample-Mill (Fuji Paudal Co., Ltd.). Ethyl cellulose (EC, 30—50 cps), cholesterol (CH, SP reagent grade) and stearyltrimethylammonium chloride (STAC) were used as purchased from Nacalai Tesque Co., Ltd. Talc (JP XI grade, Maruhitsu) and anhydrous silica (Aerosil No. 200, Nippon Aerosil Co., Ltd.) were also used as purchased.

Coating A Glatt GPCG-1 Wurster was used. A spray nozzle of 0.6 mm diameter and a laminated filter with about 1 μm opening were set throughout all experiments.

Particle Size Distribution Sieve analysis was performed. All samples were premixed with anhydrous silica of 1%. An Alpine 200LS air jet sieve was operated at a charged weight of 10 g and a sieving time of 5 min in the range of 32 to 75 μm; in a range smaller than 32 μm, microsieves of 10, 20 and 30 μm were used at a charged weight of 1 g each, and the sieving was repeated until a constant weight was reached after 2 min of operation. In a range larger than 75 μm, a row-tap shaker (Iida Seisakusho Co., Ltd.) was used; the shaking time was 10 min and the charged weight was 20 g.

Droplet Size Distribution The sizes of sprayed droplets were determined at a distance of 30 mm from the nozzle by a light scattering spray analyzer (LDSA-1300A, Tonichi Computer Applications Co., Ltd.).

Dissolution Dissolution tests were performed as previously reported.

Scanning Electron Microscopy (SEM) A Hitachi S430 was used.

Results and Discussion

Particle Design The attempt was made to finally produce as small microcapsules as possible. Since particles of 32 to 44 μm had been successfully processed and the microcapsules of 56 μm had been produced in a previous study, it was attempted here to obtain products with a
mass median diameter smaller than 30 μm.

The particle size distributions of pulverized phenacetin powders are shown in Fig. 1, where once or twice pulverized powder is respectively denoted by P_a or P_b. Mass median diameters were 19 and 11 μm, respectively, and particle sizes were well described by the log-normal distribution. Fine phenacetin particles with a low density of 1.24 g/cm³ did not seem to be discretely and steadily recycled in the Wurster chamber. Hence, the coating process was designed for fine particles to first be agglomerated at minimum until a steady fluidization was achieved and thereafter for the fine agglomerates to be coated under restrained agglomeration.

From the previous study, the membrane thickness required for practically sustained release of phenacetin was estimated to be 1.5 μm, when EC–CH–STAC (2:1:0.03) was used as a membrane material. In that case, the coating level was 6.25% for cores of 32–44 μm. Hence, nearly 75 or 25% coating should respectively be required for 11 or 19 μm cores used in this study, if the particles were to be discretely coated without agglomeration. However, slightly agglomerated particles had exhibited a several times higher release rate. A higher degree of agglomeration should not be avoided with the finer, lighter phenacetin particles, compared with calcium carbonate of 32–44 μm. In addition, the powders used in this study had a broader distribution of particle size (Fig. 1). These suggested that a stronger barrier against drug release might be required in this study.

The previous study also showed that, as the CH content became higher, phenacetin release and particle agglomeration were more suppressed. Hence, the CH content was increased to EC:CH = 1:1. To reduce excessive agglomeration which might still occur even with EC–CH (1:1), talc was used as an anticoagulant in the granulation process. Although the talc powder had a large mass median diameter of 15 μm (Fig. 1), it should be brittle and easily flaked off, and was expected to lead to suppression of the size-enlargement of agglomerates.

**Microgranulation** Composition of the spray solution and the operating conditions for the microgranulation process with two phenacetin powders are shown in Table I. The concentration of EC–CH–talc (1:1:1) spray solution was 5% (w/v) (denoted as EC plus CH below). The operating conditions were specialized by a very high spray rate. This was because the phenacetin powder was adhered to the chamber wall in large quantity due to electrostatic charging, when sprayed at lower rates. At the beginning of the operation, the powder containing a great deal of solvent was remarkably flocculated and gradually began to flow freely. The phenacetin powder was granulated to the 3, 6, 9 or 12% level (denoted below as EC plus CH relative to phenacetin) and dried until the outlet air temperature had risen by 3 °C. Granules taken from the chamber were sieved for particle size analysis.

The change in median diameter (50% diameter) of granules is shown in Fig. 2 for the two phenacetin powders of different sizes. A significant difference in the time-course of agglomeration was observed between P_a and P_b. With P_b, the particle size distribution was little changed at the beginning of granulation and the mass median diameter was suddenly increased above the 3% binder level with 5% spray solution. A similar profile was observed with the 3% solution (Fig. 2), where the delay of size-enlargement was prolonged up to the 6% binder level. On the other hand, the mass median diameter was almost linearly increased for P_b.

A profile of delayed agglomeration was reported by...
Shinoda et al. in the top-sprayed granulation of crystalline cellulose with water as a binder. In their study, the granulation delay was not observed with corn starch, lactose or mannitol. The water-soluble lactose and mannitol were very quickly granulated and the insoluble corn starch more slowly. Since water was used as a binder, lactose and mannitol dissolved in spray droplets must have acted as interparticulate bridging material. The strong water-absorption of insoluble crystalline cellulose, in contrast, caused the delay, because the water absorbed by crystalline cellulose at the beginning of the granulation process could not effectively act as a binder, different from the case of corn starch.

The results of Shinoda et al. suggested that the low coalescence probability during the lag time accounted for the delay of granulation. In the Wurster process where particles are strongly agitated, the larger the particle size is, the lower is the coalescence probability. The delay observed only with the larger \( P_s \) in this study would partly result from easier disintegration of the floculates composed of larger particles.

Sugimori et al. analyzed their experimental results from fluidized bed granulation by computer simulation. When the granulation was assumed to proceed through the generation of nuclei, the experimental results agreed well with the simulation. When the nuclei were composed of 10 or less particles, there appeared a delay of granulation. It corresponded to the case where the ratio of droplet size to particle size was smaller than 1.2. The granulation proceeded more rapidly and was less delayed as the ratio became larger.

The droplet size measurement with the 5% spray solution in this study had complicated results. About 40% of the droplets were distributed in fractions smaller than 5 \( \mu m \), and the residue was in the fraction of 8–30 \( \mu m \). The mass median diameter of the latter was 15 \( \mu m \). This suggested that the talc powder mixed into the spray solution (Fig. 1) might be causing the production of droplets of 15 \( \mu m \). Since the ratio of the droplet size of 15 \( \mu m \) to the particle sizes of 11 and 19 \( \mu m \) were 1.4 and 0.8, respectively, the sizes of sprayed droplets might also explain why the delay of granulation was observed only with \( P_s \).

In Fig. 3, the reduced mass median diameter, \( D_r/D_0 \), is plotted against the percentage of binder applied, where \( D_r \) and \( D_0 \) are the mass median diameters of granules and raw powders, respectively; with \( P \) the binder applied is made zero at the starting point of rapid size-enlargement. The data were well fitted to one line. This showed that granulation proceeded at the same rate for both powders and both concentrations in the case of \( P_s \). However, in the granulation process, longer delay is usually followed by slower agglomeration, as well demonstrated by the simulation of Sugimori et al. and the experimental results of Sagawa and Sakamoto and Shinoda et al. According to their results, the granulation in this study would have proceeded faster with \( P_s \) than with \( P \) and faster at 5% than at 3%.

In the present work, the difference of granulation between \( P_s \) and \( P \) in the Wurster process cannot be clearly explained. The strong agitation which acted on particles and agglomerates might have complicated the particle growth. Although \( P_s \) and \( P \) exhibited the same granulation rate after the delay in the experimental range used, the granulation of \( P_s \) even seemed to have a plateau after a longer granulation (Fig. 3). The complicated size distribution of the droplets containing solid particles also has to be further studied. Studies on elucidation of the mechanisms will be reported in the future.

As a model of fine powder, \( P_s \) was adopted below, because the particle size of \( P_0 \) (Fig. 1) seemed too large and its agglomeration consequently led to the production of granules that were too coarse (Fig. 2).

Figure 4 shows the effect of drying in the chamber on the change of mass median diameter of \( P_s \) granules. The concentration of spray solution was fixed at 5% as EC plus CH. When the three kinds of formulations used were compared, talc was seen to be effective in restraining size-enlargement, though only a little. On the other hand, the difference of mass median diameter between undried and dried samples was very large at 3 and 6% coating levels, becoming 6 and 9 \( \mu m \) (EC–CH–talc), respectively. These were very large compared with the desired size of the final product. These results suggested that the disintegration of weakly coagulated particles by drying prior to
coating might be effective in reducing the particle size of microcapsules.

The granulation process has been studied in detail by Sekiguchi and Oota.\textsuperscript{9}) Granulation proceeds through the nuclei growth, transition and pellet growth (secondary agglomeration) regions. Apparently the granulation process demonstrated with the wet sample (undried) in Fig. 4 corresponded to the nuclei growth region. Sekiguchi and Oota\textsuperscript{9}) expressed the nuclei growth by Eq. 1, where the decrease in particle number was assumed to result from the coalescence and the destruction described by the second and first order kinetics, respectively.

\[ \frac{dN}{dt} = -AN^2 + BN \]  

(1)

where \( A \) and \( B \) are the rate constants and \( t \) the amount of binder applied (\%). Equation 1 gave Eq. 2 by integration.

\[ \ln \frac{D_{x,0} - D_{x,t}}{D_{y,0} - D_{y,t}} = -Bt \]  

(2)

where \( D_{x,0} \), \( D_{x,t} \) and \( D_{y,0} \) are the volume mean diameters at \( t = 0 \), \( t \) and \( \infty \), respectively. \( D_{x} \) can be replaced by the mass median diameter \( (D_{50}) \), when the particle size distributions during granulation are be described by the normal or log-normal probability law and their standard deviations are constant.

The size-enlargement of wet granules (Fig. 4) seemed to reach an equilibrium, as reported by Sekiguchi and Oota.\textsuperscript{9}) The particle size distributions of the wet granules were described on the log-normal probability paper by the lines broken at 32 \( \mu \)m; however, they seemed to be roughly approximated by the same standard deviation as that of \( P_b \). By the least squares method based on Eq. 2 replaced \( D_{x} \) by \( D_{S0} \), \( D_{50} \) (\( \mu \)m) could be expressed by Eq. 3 for the wet granules prepared with EC–CH–talc.

\[ D_{S0} = 51.15 - (51.15 - 10.89) \exp (-0.0257t) \]  

(3)

The decrease in mass median diameter by drying clearly showed that weakly agglomerated particles still remained (Fig. 4). When the granule size would no longer be reduced by drying, the nucleation seemed to reach the equilibrium state, where the granules would become compact enough to accept an efficient coating. At the intersecting point of the linear regression line for dried granules (Fig. 4) and Eq. 3, the binder applied was 21.3\% and the mass median diameter 38.5 \( \mu \)m. Granules of 38.5 \( \mu \)m were not available in this study which was aimed at producing small microcapsules, though they might be found useful for other applications.

The change of particle weight in each size fraction during granulation with EC–CH–talc is shown in Fig. 5 for dried granules. The weight of particles smaller than 10 \( \mu \)m was predominantly decreased, while that of particles larger than 20 \( \mu \)m was monotonously increased. The weight of 10–20 \( \mu \)m particles was at first increased due to the fast granulation of those smaller than 10 \( \mu \)m. It then decreased above the 6\% binder level, which suggested that the 10–20 \( \mu \)m particles were also agglomerated. The production of particles larger than 53 \( \mu \)m was curtailed below 10\%. The behavior of the 78\% particles smaller than 20 \( \mu \)m which were contained in \( P_b \) played an important role in the granulation.

**Microencapsulation with EC–CH–STAC (1:1:0.06)**

Details of the cores, the composition of spray solution and the operating conditions for both granulation and coating are listed in Table II. The spray solution of 5\% and the phenacetin powder of \( P_b \) were used. At the beginning of the coating process, the operating conditions were changed to achieve a drier state and more rapid particle motion, and thus to suppress the agglomeration. For that purpose, the spray pressure and inlet air rate were increased to 3.5 at and 0.8 atm and 0.8 m\(^3\)/min, respectively; the spray rate was decreased to about 2.8 ml/min. Talc was removed from the coating solution, since it had a large particle size of 15 \( \mu \)m, potentially resulting in formation of the particles with irregular, rough surface. Instead of talc, STAC was mixed to reduce particle adhesion due to electrostatic charge.\textsuperscript{1)}

In experiment-1 (exp.-1), the binder level in the granulation process was made 3\%, where the powder seemed to achieve a free-flowing state. Wet granules (19 \( \mu \)m) were dried up to 3\% C higher outlet air temperature to reduce their size to 14 \( \mu \)m and thereafter coated at 11.3 ml/min.

| Table II: Operating Conditions in Granulation and Coating |
|----------------------------------|------------------|------------------|------------------|
|                                  | Granulation      | Coating          | Granulation      | Coating          |
|                                  | Raw material: phenacetin, \( P_b \) (11 \( \mu \)m) | Charged weight (g) | 250                      | 250                      |
| Spray solution:                  |                  |                  |                  |                  |
| EC                               | 3.75 g           | 75 g             | 3.75 g           | 75 g             |
| CH                               | 3.75 g           | 75 g             | 75 g             | 75 g             |
| Talc                             | 3.75 g           | 75 g             | 75 g             | 75 g             |
| STAC                             | 4.5 g            | 4.5 g            | 4.5 g            | 4.5 g            |
| Ethanol                          | 300 ml           | 300 ml           | 300 ml           | 300 ml           |
| Operating conditions:            |                  |                  |                  |                  |
| Inlet air temperature (\( ^\circ \)C) | 25–29            | 25–29            | 25–29            | 25–30            |
| Outlet air temperature (\( ^\circ \)C) | 0.2              | 0.2              | 0.2              | 0.2              |
| Inlet air rate (m\(^3\)/min)    | 14.2             | 14.2             | 14.2             | 10.9             |
| Spray rate (ml/min)              | 2.5              | 2.5              | 2.5              | 3.5              |
| Size of the product\( p \) (\( \mu \)m) | 14               | 31               | 27               | 44               |
| Yield (%)                        | 90               | 90               | 90               | 90               |
| Symbol of microcapsule           | MC-1             | MC-1             | MC-2             | MC-2             |

\( p \) Mass median diameter.

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**Fig. 5.** Variation of the Weight of Dried Granules in Size Fractions during Granulation of \( P_b \), with EC–CH–Talc (1:1:1)

Size fraction: \( \bullet \), 10–20 \( \mu \)m; \( \triangle \), 20–32 \( \mu \)m; \( \Delta \), 32–53 \( \mu \)m; \( \square \), + 53 \( \mu \)m.
The mass median diameter of the product was 31 µm at 60% coating level. The final yield was 90%.

As a reference, coating was performed under the same conditions without drying the wet granules, after the granulation was performed up to 6% binder level. Hence, at the beginning of the coating process, the mass median diameter of wet particles recycling in the chamber was 27 µm (Fig. 4). In this case, the mass median diameter of the product reached 44 µm at 60% coating level (Table II). In both exp.-1 and 2, there occurred a similar increase of 17 µm in the mass median diameter during coating.

In general, the agglomeration rate is strongly dependent on the sizes of sprayed droplets. Schaefer and Worts\(^\text{9}\) reported that the droplet size was dominated by air-to-liquid mass ratio at a constant concentration. Therefore, at a constant spray pressure, a lower spray rate should result in smaller droplet size and, consequently, in lower agglomeration due to weakening of the interparticulate bridge or decrease in the size of nuclei.\(^\text{9}\) To reduce the agglomeration encountered in exp.-2, the spray rate was decreased to 6.2 ml/min. However, the mass median diameter of the product was contrarily increased to 99 µm. This can be explained by the view that the low spray rate led to the adhesion of small particles to the chamber wall due to the electrostatic charging, the coalescence of the oversprayed large particles and the layering of the small particles on large particles predominantly sprayed.\(^\text{4}\)

The change of mass median diameter throughout the process is shown in Fig. 6. The wet sample was withdrawn at each coating level, mixed with 1% anhydrous silica and sieved without drying. In both exp.-1 and 2, the particle size was gradually increased until the 60% coating level. Comparison of exp.-1 and 2 revealed a difference only in the size of granules. The initial difference in granule size remained almost constant during coating. The granulation in exp.-1 reduced to the 3% level and the drying prior to coating were effective in decreasing the particle size of the product.

The particle size distributions of the products are shown in Fig. 7, compared with those of the related powder and granules. The product from exp.-1 (MC-1) was characterized by smaller mass median diameter and broader distribution. Although the product from exp.-2 (MC-2) had a large mass median diameter of 44 µm, its particle size distribution was sharper and the amount larger than 106 µm was not significantly different from that of MC-1.

The increase in particle size should continue until the smallest particles in the system became no longer agglomerated. The change of particle weight in each size fraction during coating is shown in Fig. 8. In both exp.-1 and 2, particles smaller than 20 µm were reduced in number and seemed to finally disappear; this is most clearly demonstrated by the results from exp.-2. The smallest size of the microcapsules coated as discrete particles without agglomeration was nearly 20 µm under the coating conditions set in this study (Table II). This lower particle size limit (20 µm) should be clearly dependent on the exerted agitation, the strength of interparticulate bridging materials and the droplet size. The coating conditions used in this study (Table II) were the strongest limit of agitation to be exerted on particles. The droplet size was also the lowest limit, since greater spray pressure caused the ejection of wet particles and droplets to the filter and a lower spray rate.
caused particle adhesion to the wall due to electrostatic charging. Hence, if finer microcapsules from the Wurster process were desired, membrane materials of lower strength would have to be developed.

Sugimori et al.\textsuperscript{7} reported the relation of the mean nucleus size $NN$ (the number of particles in a nucleus) to the mean droplet size $D_o$ in the granulation (Eq. 4).

$$NN = 6.44(D_o/D_p)^3$$

where $D_o$ is the mean particle size. The experiments of Sugimori et al.\textsuperscript{7} were limited only to top-sprayed, fluidized bed granulation with a 5\% hydroxypropyl cellulose aqueous solution. Although the analysis should be based on more comprehensive granulation processes, the difference in the agitation and the strength of membrane material or binder would roughly be reduced to that in the nucleus number $NN$. The size distribution of the droplets of EC–CH–STAC (1:1:0.06) solution sprayed in this study (Table II) was well explained by the log-normal probability law. The mass median diameter was 8.3 $\mu m$ and the geometric standard deviation ($\sigma_g$) 1.79. Hence, it was estimated from Eq. 4 that the particles which could be granulated by the nucleation of two particles (the smallest nucleus size) might be 12 $\mu m$ in diameter. Since this size of 12 $\mu m$ was a mean diameter of core powder, the above estimation might explain the result that the lowest size of the particles to be coated without agglomeration was about 20 $\mu m$.

The dissolution profiles for MC-1 and 2 are shown in Fig. 9. The results showed 40\% or more coated products to be practically useful for sustained release. The faster release from MC-1 than from MC-2 clearly resulted from its smaller particle size.

In Fig. 10 are shown SEM photographs of MC-1, which well demonstrate the process of size-enlargement. The granules (a) already contained coarse particles and fine particles adhered to the granular surfaces (Fig. 10a). After 20\% coating (b), the fine particles seemed to disappear as expected from Fig. 8, but secondary agglomerates with irregular shape and porous structure were observed. Such insufficient encapsulation clearly led to the fast dissolution from 20\% coated microcapsules (Fig. 9). At 60\% coating level, the surfaces of particles became smooth and the pores became well covered with membrane (Figs. 10c and d). The fine particles around 20 $\mu m$ were well coated at 60\%, though the poor encapsulation of several percent of them might account for the bursting at the beginning of dissolution (Fig. 9).

**Conclusion**

A phenacetin powder with a mass median diameter of 11 $\mu m$ was slightly granulated up to 19 $\mu m$ using 3\% binder, dried to reduce the size to 14 $\mu m$ and subsequently coated by the Wurster process. The microcapsules produced had a mass median diameter of 31 $\mu m$ at 60\% coating level and exhibited a practically useful sustained release at 40\% or above.

The agglomeration in the coating process seemed to continue until the smallest particles in the system were no longer agglomerated. The smallest size of the particles which could be processed with no successive agglomeration was estimated to be 20 $\mu m$. This would be the smallest microcapsules that the Wurster process has ever produced. To lower this limit further, the membrane material must be much weaker than EC–CH–STAC (1:1:0.06).

On the other hand, the agglomeration of particles smaller than the 20 $\mu m$ current minimum led to the production of coarse particles, which accounted for a broad particle size distribution of product. The removal of particles smaller than the limit from raw powder would be effective for producing single-core microcapsules of the order of 20 $\mu m$.

**Acknowledgements** The authors would like to thank the staff of the Department of Research and Development, Okawara Seisakusho Co., Ltd., for their technical assistance in the measurement of droplet sizes.
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