Optimization of Experimental Conditions for the Compression and Stress Relaxation Test of Spray-Dried Granules

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1. Introduction

In the press forming process of ceramics, it is important to accurately characterize the mechanical properties of spray-dried granules. In many researches on the characterization of granules, the mechanical properties were often evaluated by applying the conventional compression test, in which the packing density of granule bed was measured as a function of applied pressure, or the single granule compression test, in which the strength of a granule was measured. These methods give us information only on the density of green body, but not on the homogeneity of green body.

In pharmaceutical science, the static compression behaviors and the tablettabilities of the agglomerated crystals and the original crystals were investigated and compared by measuring the stress relaxation. In this report it was found that the relaxation pressure of the agglomerated crystals was higher than that of the original crystals. It was also shown that the tensile strength of the tablet of agglomerated crystals was greater than that of the original crystals. However several kinds of agglomerated powders were not compared, therefore it is not clear the difference of the breaking and/or deforming behavior of agglomerated powders and how to prepare the optimum agglomerated powders for the press forming. Furthermore the effect of the experimental conditions on the stress relaxation behavior has not been discussed sufficiently. In food science, compression and stress relaxation tests have been used for the textual evaluation of various fruits and other food commodities. Therefore we need a few hours to proceed a single run and the optimization of the experimental conditions hasn’t been done yet. In the present study we discuss the dependency of the compression rate and the initial height of the granules bed on the relaxation ratio in order to obtain the optimal conditions which reasonably evaluate the stress relaxation behavior of granules. Furthermore in order to shorten the experimental time, reducing the compression number and the relaxation time are undertaken.

2. Experimental

2.1 Granule preparation

Two kinds of slurries were prepared from commercial-grade \(\alpha\)-\(\text{Al}_2\text{O}_3\) powders (AL-160SG-3, Showa Denko) and distilled water by ball-milling for 8 h using alumina balls in an alumina bottle. Solid content was 27 vol\%. Polycarbonate ammonium was used as a dispersant. After 8 h ball-milling, one of slurries was added with the binder and an additional 2 h ball-milling. All the granules were prepared from these slurries under the same conditions for spray-drying. The details of slurry preparing conditions are summarized in Table 1, in which the morphology of the spray-dried granules was also mentioned.

2.2 Compression and stress relaxation test

2.2.1 Multiple compression and stress relaxation test

The granules, sieved within a range of 45–63 \(\mu\)m and packed with an initial height of 2 mm, were pressed in a die (inner diameter 16 mm) mounted on the stage of a standard
mechanical testing machine (SDW-2000, Imada Co.). The granules were compressed 0.15 mm in depth and then relaxed for 15 min in each step. This compression and relaxation procedure was repeated until the final compression pressure of 90 MPa. The compression rate was controlled at 0.04, 0.14 and 1.2 mm/min in order to evaluate the dependency of the compression rate on the stress relaxation.

2.2.2 Single compression and stress relaxation test

In order to shorten the experimental time the compression number was reduced to one time. The granules were compressed until the applied pressure of 1 MPa at compression rate of 1.2 mm/min, and then the piston was stopped. The relaxation pressure was measured as a function of the time after compression halting. Sample granules and packing procedure of granules were same as the multiple compression and stress relaxation test (2.2.1). Furthermore the single compression and stress relaxation test was done while the initial height of granules bed was controlled at 2, 4 and 7 mm.

3. Results and discussion

3.1 Dependency of the compression rate

Figure 1 shows the schematic illustration of a compression and stress relaxation curve typically observed at each step. This stress relaxation behavior can be divided into three parts: the pressure which dropped suddenly after compression halting (termed plastic relaxation), the pressure which gradually decreased (viscous relaxation), and the residual pressure after the relaxation process (elastic residue). The plastic relaxation is ascribed to the release of the elastic energy at granules' contacting points, caused by granule fracture. The viscous relaxation is ascribed to the release of the elastic energy at granules' contacting points, caused by granule deformation or microscopic granule rearrangement. The elastic residue is mainly attributed to the elastic energy at granules' contacting points. Since the rate of plastic relaxation is low to be clearly distinguished, the plastic relaxation is included in the viscous relaxation and represented as a relaxation, \( P_{p} - P_{e} \). The relaxation \( P_{p} - P_{e} \) is divided by the applied pressure \( P_{p} \) and represented as a relaxation ratio, \( (P_{p} - P_{e})/P_{p} \).

The transient behavior of stress relaxation is shown in Fig. 2 (a), where the relaxation ratio, \( (P_{p} - P_{e})/P_{p} \) is plotted as a function of packing fraction. Fig. 2 (b) shows the relaxation ratio at packing fraction of 0.38. For both G3 and G16, the value of the relaxation ratio increase with an increase in the compression rate. In order to discuss the dependency of compression rate on the relaxation ratio qualitatively, we introduce a simple rheological model where the elastic deformation at granules' contacting points is represented by springs and the granules' microscopic rearrangement and deformation by dashpots. The Maxwell model in which a...
spring and a dashpot are connected in series can't express
the elastic residue, therefore we applied the three elements
model shown in Fig. 3. In this model, the reaction pressure
of the dashpot increases with increasing compression rate.
This reduces the strain of the dashpot and increases the
strain of the spring $k_2$. As the compression rate increases,
the elastic energy stored by the spring $k_2$ increases during
compression, and the amount of the stress relaxation stem-
ming from the dashpot increases.

From the above results, the difference of the relaxation
ratios between G3 and G16 should appear more clearly
when the compression rate is high. In other words, a high
compression rate should be used to evaluate the granule vis-
cous deformation.

3.2 Single compression and stress relaxation test

The relaxation ratio has the maximum value at the pack-
ing fraction 0.35–0.40, as shown in Fig. 2. Figure 4 shows
the relationship between the packing fraction of the granule
bed $\phi_c (=1-\varepsilon_c)$ and the inter-granule porosity $\varepsilon_g$. The inter-
granule porosity $\varepsilon_g$ was calculated by the following equation,

\[ (1-\varepsilon_g)(1-\varepsilon_i) = 1-\varepsilon_c \]

where $\varepsilon_i$ is the intra-granule porosity (-). The intra-gran-
ule porosity $\varepsilon_i$ was measured by mercury porosimetry
(Table 1). Assuming that granules are spherical, $\varepsilon_g$ is 0.26
at the closest packing of granules. In Fig. 4 it is shown that
the closest packing of granules was attained at the packing
fraction of granule bed 0.35–0.40. This results explain that
the stored elastic energy of granules was maximum at the
packing fraction 0.35–0.40, resulting in the maximum of the
relaxation ratio. We can reduce the compression number to
one time if the granule bed was compressed until the pack-
ing fraction 0.35–0.40 for the measurement of stress relaxa-
tion, and the relaxation ratio was compared between G3 and
G16. However it was hard to estimate the packing fraction
of granule bed during the test because we had to take out
the sample and weigh for the estimation. If the relationship
between the packing fraction and the applied pressure is
known before the test, we can control the applied pressure
instead of packing fraction. The relationship between the
packing fraction and the applied pressure can be obtained
by the conventional compression test.\(^\text{1-3}\) The applied pres-
sure was determined to be 1 MPa because the pressure cor-
responding to the packing fraction 0.35–0.40 is 1 MPa, as
shown in Fig. 5, the results of the conventional compression
test. Figure 6 shows the change of the relaxation ratio after
compression of 1 MPa, where the relaxation ratio plotted as
a function of the time after piston halting. The difference of
the relaxation ratio between G3 and G16 can be clearly dis-
tinguished by the single compression and stress relaxation
test, showing the good agreement with those obtained from
the multiple compression and stress relaxation test (Fig. 2).
It is also shown that the relaxation time can be shorten to
about one minute in order to evaluate the relaxation ratio.

3.3 Influence of the initial height

Figure 7(a) shows the change of the relaxation ratio
measured under three kinds of initial height, 2, 4 and 7 mm.
In Fig. 7(b) the values of the relaxation ratio at 8 min after
piston halting are compared. For both G3 and G16, the
values of the relaxation ratio are almost independent of the initial height of the granule bed. Generally the effect of the wall friction increase with an increase in the initial height of granule bed, resulting in insufficient transmission of the compression pressure. However during the stress relaxation process, the displacement of granules should not be occurred and there should be no effect of the wall friction, resulting in the almost same values of the relaxation ratio.

4. Conclusions
Two kinds of spray-dried granules were subjected to the multiple compression and stress relaxation test and the single compression and stress relaxation test. The compression rate and the initial height of the granule bed were controlled and compared the relaxation ratio. From the above experiments, the followings are concluded.

(1) The relaxation ratio increases with an increase in the compression rate. It is shown that the difference in the value of the relaxation ratio should appear clearly when the compression rate is higher.

(2) The relaxation ratio obtained from the single compression and stress relaxation test is a good agreement with those obtained from the multiple compression and stress relaxation test. This result can shorten the total experimental time from a few hours to about ten minutes.

(3) The relaxation ratio is almost independent of the initial height of the granule bed.

References