Development of a Novel Compression Tester and Rheo-Mechanical Properties of Wet-Mass Powder. I
—Effect of Kneading Time on the Rheo-Mechanical Properties—

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In our previous paper [Watano S., *et al.*, *Chem. Phram. Bull.*, 49(1), 64—68, (2001)], a compaction tester was developed to quantitatively evaluate the water dispersion condition of wet kneaded masses prepared by a paddle type kneader. It was also demonstrated that the physical properties of pellets prepared by extrusion granulation after the kneading could be well predicted by the vertical pressure transmission obtained through the compaction tester. However, in this compression tester, the vertical pressure transmission was just obtained and rheological and mechanical properties (so called rheo-mechanical properties) of wet mass-powder that should be the most important to determine the deformation process were not well studied. In this study, a novel compression tester, which can measure both vertical and radial pressure transmissions, has been developed. Based on the compression test, mechanical property (Young's modulus) and rheological property (effective internal friction) of wet mass powder prepared by different kneading times were quantitatively investigated. Granules (pellets) were then obtained through the extrusion granulation and fluidized bed drying, and the physical properties (strength and disintegration time) of the obtained pellet were evaluated. The relationship between the granule (pellet) physical properties and the mechanical and rheological (rheo-mechanical) properties was analyzed.

Key words compression tester; wet-mass powder; rheo-mechanical property; deformation; kneading

Extrusion process has widely been used in the pharmaceutical and agriculture formulations, foods, ceramics, chemicals and other industries, making use of its advantages that the construction and operation are simple, mass production is easy with low cost, and uniform physical properties (size, density, main ingredient content and *etc*.) can be obtained.

Up to this time, many studies have been made on the extrusion process. For the process monitoring, measurement of extrusion pressure has been used to understand the extrusion process.[2,3] It was reported that it could be used to estimate the extruder performance.[4] Studies on the effect of the process parameters such as particle size,[5] mixing procedure[6] and kneading condition[1] on the process have also been conducted. In our previous study,[1] we have developed a compression tester to measure the vertical pressure transmission among a wet kneaded mass and have demonstrated that the vertical pressure transmission could predict the water dispersion condition among the mass, the degree of kneading attained, and properties of dry pellets prepared by extrusion granulation after the kneading. However, a practical method, which can probe the rheological and mechanical properties (so called rheo-mecahnical properties)[7] and micro-structural characteristics of wet-mass powder, has not been proposed yet, although they are very important to determine the final product attributes and the process conditions. Also, the deformation process of wet-mass powder during the compression has not been discussed in detail.

In this study, a novel compression tester with vertical and radial pressure transmission measurements was developed and a practical method to probe rheo-mechanical properties of wet-mass powder was proposed. The obtained method was then applied to the actual extrusion operation, and its validity was investigated. Relationship between the rheo-mechanical properties of wet-mass powder and the physical properties of dry extruded pellets was also investigated. The deformation process during the compaction was also discussed here.

Experimental

Powder Samples Table 1 lists powder samples used.[1] Pharmaceutical excipient consisted of lactose, cornstarch and crystallinecellulose, and dry hydroxypropylcellulose (HPC-L) as binder were mixed together, and then kneaded with purified water for predetermined time. The water content was fixed at 25% constant.

Equipment and Procedures For wet kneading, a five litter planetary mixer (Twin-servo, Shinagawa Manufacturing Co., Ltd.) was used. The

Table 1. Powder Samples

Samples	Number median diameter (μm)	Charge mass ratio (%)	Charge mass (kg)
Lactose	60	67.2	0.672
Corn starch	15	28.8	0.288
Crystallinecellulose	40	4.0	0.040
Total		100.0	1.000
Hydroxypropylcellulose (HPC-L)	21	3.0[a)]	0.030
Purified water		22.0[a]	0.220

a) Binder (HPC-L) and binder liquid (purified water) are mixed to 1.0 kg of the excipient.

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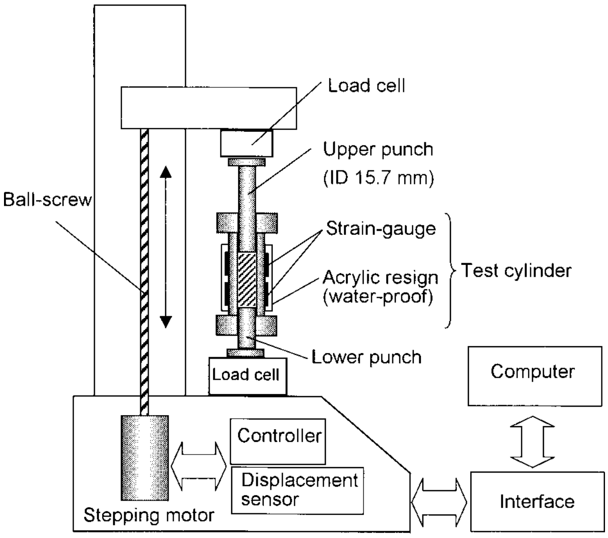

Fig. 1. Schematic Diagram of Compression Tester

kneading chamber has two top-drive paddles, each of which revolves while tuning round on its axis in a different speed (revolution and turning speeds are 10 rpm and 4.025 rpm, respectively).

A kneading experiment was conducted as follows: powder samples and dry binder (HPC-L) were fed into the kneading chamber and mixed for 2 min. Then the mixture was kneaded with purified water for predetermined time. After the kneading, the kneaded wet-mass powder was extruded through an extrusion granulator (DG-L1, Fuji Paudal, Co., Ltd.), which consisted of a hemispherical dorm type punching screen (diameter of the dome was 58 mm, diameter of each hole and width of the screen were both 0.8 mm, and the opening area ratio was 22.5%), a single shaft with screws and an extrusion blade at the extremity of the shaft.[1,8] The dry product (pellets) was finally obtained after a fluidized bed drying (NQ-125, Fuji Paudal, Co. Ltd.)[9] of wet extruded pellets under the air temperature of 353 K. Physical properties of the dried pellets were evaluated by their strength and dissolution time (the method for measuring the granule strength and dissolution time was the same as previously reported[1]).

A compression test of the kneaded mass was conducted using a developed compression tester (Fig. 1). The tester consists of a stepping motor with ball screws, displacement sensor, upper and lower punches and load cells, test cylinder, and computer. The maximum load is 5000 N (50 MPa at 1 cm^2) and a personal computer with MS Windows software is installed to continuously measure the pressures and displacement, as well as automatically operate the compression test.

As shown in the figure, the cylinder has a dimension of 110 mm long with ID of 15.7 mm (cross sectional area is just 2.0 cm^2) and wall thickness of 1 mm. Four sets of strain gauges are glued on the outside wall of the cylinder, which accurately measure radial load. The upper punch presses the wet-mass powder at the moving speed of 50 mm/min and the pressure transmission (G), Rankin factor (K) and effective internal friction (tan ϕ) can be computed automatically.

The pressure transmission, G, was calculated by using the following equation (Eq. (1)).

$$G=P_L/P_U\times 100 \tag{1}$$

where P_L and P_U indicate the pressure of the lower and upper punches, respectively.

In general, pressure transmission among the powder bed can be expressed by a Shaxby–Unkel equation as,[10]

$$G=\exp\left(\frac{-4\mu KH}{D}\right) \tag{2}$$

where H, D and μ indicate height of powder bed, diameter of cylinder and external friction, respectively.

The Rankin factor, K, shown in the Eq. (2) can be written by radial pressure P_D subjected to the upper pressure P_U as

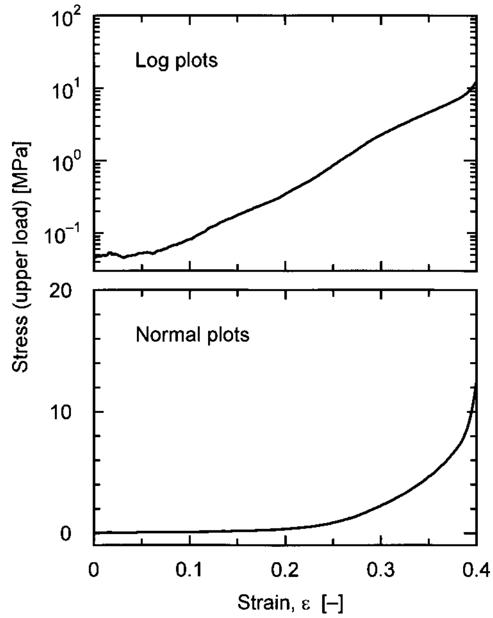

Fig. 2. Conventional Stress-Strain Diagram of Wet-Mass Powder

$$K=\frac{P_D}{P_U} \tag{3}$$

The effective internal friction can be described as tan ϕ, assuming the angle of internal friction is ϕ. This ϕ can be obtained by the Rankin factor K as follows.

$$K=\frac{1-\sin\phi}{1+\sin\phi} \tag{4}$$

For the elastic deformation region, Young's modulus, E, can be calculated by the Eq. (5)

$$F=E\cdot\varepsilon \tag{5}$$

where, F and ε indicate load and strain, respectively.

In this study, the rheo-mechanical properties of wet-mass powder are evaluated by the Young's modulus (E), pressure transmission (G), effective internal friction (tan ϕ), and critical point (P_S) which will be described later. All of the data.

Results and Discussion

Compression Diagram Figure 2 confirms stress-strain diagram of wet mass powder by using normal and log plots. Seen from this figure, the stress-strain diagram is totally different from the conventional diagram; it is difficult to find out the elasticity limit, yield point and *etc*. Therefore, the conventional stress-strain diagram cannot be used for the wet system, due to the complicated wet-mass powder's characteristics, *i.e.*, non-continuous with multi phases (gas, solid and liquid). We here propose to use pressure transmission diagram to analyze physical properties of wet-mass powder.

Figure 3 investigates the relationship between pressure transmission (G) and stress (upper load, P_U). Seen from the figure, the pressure transmission-stress diagram can be divided by four phases. The mechanical characteristics of each phase can be analyzed as follows.

(Phase I) Re-arrangement of wet mass powder is occurred inside the cylinder. When the stress reaches point A, the elastic deformation begins.

(Phase II) The elastic deformation (relationship between stress and strain is linear) is observed. At the point B (pro-

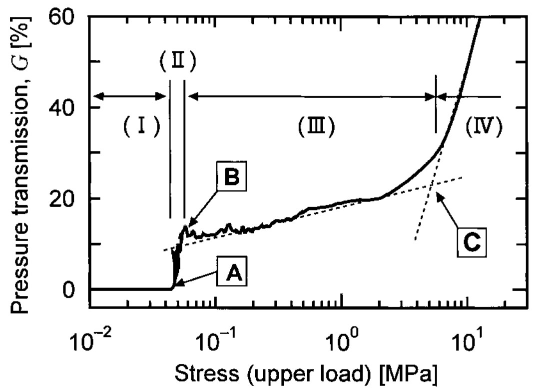

Fig. 3. Compression Diagram

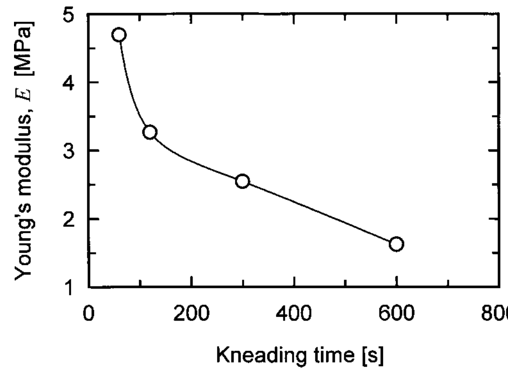

Fig. 5. Temporal Change in Young's Modulus

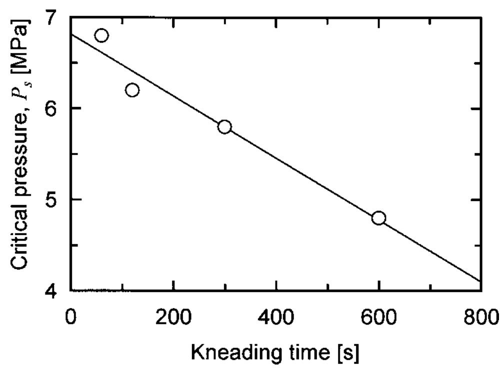

Fig. 4. Relationship between Critical Pressure (P_S) and Kneading Time

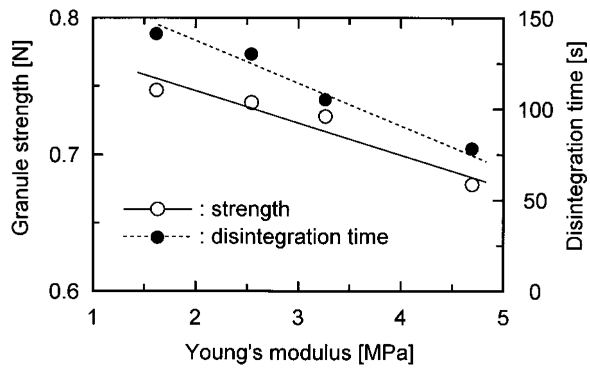

Fig. 6. Granule Physical Properties *v.s.*, Young's Modulus

portional limit), the void fraction of powder bed reaches 0, indicating the wet-mass powder can be treated as continuous.

(Phase III) The plastic deformation is observed. At the point C (stress at this point is referred to as critical pressure, P_S), the line inclination changes awfully. Before the point C, binder water is locally isolated (not uniformly distributed) and the wet-mass powder still contains gas (air), therefore, the wet-mass powder reveals complicated three-phase behavior. Due to the water isolation, pressure (upper load) transmits mainly through the solid powder, thus the vertical pressure transmission is small.

In the real calculation, the approximate straight lines are calculated for both Phase III and Phase IV by using a least square method and then point C can be obtained by the intersection of the two lines. It is noteworthy that the linearity is very well for the approximate lines and the accuracy of determination in the point C is very high.

(Phase IV) After the point C, gas (air) is removed completely and water is uniformly distributed among the powder due to the compression, showing the solid-liquid two phases behavior. Due to the water uniform distribution, pressure transmits mainly through the water. This is considered to be the main reason that the vertical pressure transmission increases greatly after the point C.

Figure 4 investigates the relationship between critical pressure and kneading time. It is obvious that the critical pressure shows a linear relationship with the kneading time. This implies that the proposed critical pressure clearly indicates the deformation characteristics of wet-mass powder prepared by the different kneading times; when the kneading progresses with longer kneading time, gas (air) is well removed from the wet-mass powder and the deformation characteristic improves (easy to be deformed when subjected to the vertical pressure), resulting in the smaller critical pressure. In other words, small upper load is large enough to obtain the plastic deformation. The critical pressure thus directly indicates deformation characteristics.

In the following, several parameters such as Young's modulus, pressure transmission (G) and effective internal friction ($\tan \phi$) can be calculated at the critical pressure. Also, relationship between these parameters and properties of dry granules (pellets) prepared by extrusion and fluidized bed drying are investigated.

Mechanical Properties of Wet Mass Powders Figure 5 shows temporal change in Young's modulus. Seen from Fig. 5, Young's modulus decreases with kneading time, showing the amount of elastic deformation increases (wet-mass powder becomes soft) due to the kneading. Basically, the plastic deformation when the wet-mass powder goes through the extrusion screen may determine the granule physical properties. Assuming that there is an analogy between the elastic and plastic deformation characters, the Young's modulus can be the indicator of predicting the granule physical properties.

Figure 6 confirms the above assumption. The relationship between Young's modulus and granule physical properties (strength and disintegration time) shows clear linearity. The smaller the Young's modulus (softer the wet-mass powder), the stronger the granule physical strength becomes. From these findings, the mechanical properties of wet mass powder are found to directly determine the granule physical properties.

Figure 7 indicates the relationship between the pressure

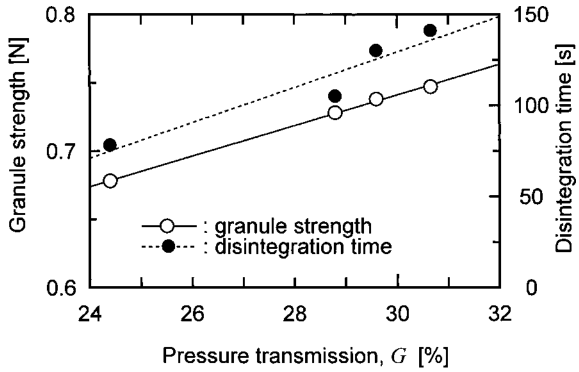

Fig. 7. Relationship between Pressure Transmission and Granule Physical Properties

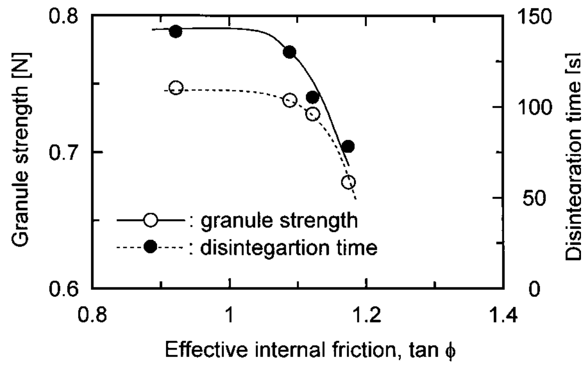

Fig. 8. Relationship between Effective Internal Friction and Granule Physical Properties

transmission and granule physical properties. Both the granule strength and disintegration time show linear relationship with the pressure transmission. The pressure transmission characteristic, an indicator of kneading condition by utilizing water dispersion condition,[1] can also be used to predict the granule physical properties. It is noteworthy that in our previous report[1] the pressure transmission was calculated based on the constant upper load (4.9 MPa), since there was no method to analyze the deformation process as shown in Fig. 3. With this previous method, it was impossible to carry out the precise analysis. However, the present method can understand the mechanical characteristics well, resulting into the precise analysis of wet-mass powder.

Rheological Properties of Wet Mass Powders Figure 8 shows the rheological properties of wet-mass powder by using the analysis of the effective internal friction, tan ϕ. By kneading the wet-mass powder under the same moisture content, longer kneading time results in the better water distribution. This improves the deformation characteristics and also decreases the particle–particle interaction (friction). In this case, the effective internal friction becomes small. Seen from the Fig. 8, no linearity can be seen between the effective internal friction and granule physical properties. However, it is obvious that the smaller the internal friction, the stronger the granule strength becomes. From these findings, it can be concluded that the mechanical properties rather than the rheological properties can affect the granule physical properties (granule strength and dissolution time) when the moisture content is the same.

In the next paper, we will investigate the effect of moisture content on the rheo-mechanical properties of wet-mass powder.

Conclusions

A novel compression tester, which can measure both vertical and radial pressure transmissions, has been developed. Quantitative analysis of rheo-mechanical properties of wet-mass powders prepared by different kneading times with the same moisture content was conducted. Extrusion granulation was then conducted and relationship between granule (pellet) physical properties and the rheo-mechanical properties was analyzed. It was found that the mechanical property (Young's modulus) and vertical pressure transmission have linear relationship with granule physical strength and dissolution time. The rheological property (effective internal friction) was also found to affect granule properties, however, no linearity between both parameters could not be obtained. It is obvious that the developed tester can probe the rheo-mechanical properties of wet-mass powder and also predict granule physical properties accurately. In the next paper, we will investigate the rheo-mecanical properties of wet-mass powders prepared by different moisture contents.

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