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Viscous Behavior of Powder in the Process of Dynamic Compression[1]

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To clarify the viscous behaviors of powder when sample which had been compressed once was recompressed, the relation between compression stress and displacement of upper punch was studied. There appeared a hysteresis loop in the stress-displacement diagram and from the loop area loss energy depending on the viscosity of sample was calculated, and the relation between loss energy and compression time was investigated. Some informations on the influence of additives upon loss energy were also obtained. It was proved that the fact that in the process of tabletting larger compression force was required when compression velocity was increased was the result of increasing of loss energy based on the viscosity of sample.

In the previous paper, the authors have analyzed the process of dynamic compression of powder, and have devised from the behaviors of compression stress and strain a new method of drawing curves which were in accord with normal creep curves obtained from the process of static compression. The authors have also clarified and reported in detail[3] by analyzing the creep curves how far plastic deformation and viscoelastic deformation contributed to the change of packing structure of powder under dynamic compression.

It was found that the time when compression stress became maximum did not agree with the time when compression strain became maximum, and there was the phase lag in the compression strain. The phase lag is the result of viscosity of sample, and the quantitative analysis of the viscous behavior is now thought to be of much importance to analyze the process of dynamic compression of powder. Because it is rather hard to elucidate precisely the viscous effect in the process of compression of powder, so presently the authors examined the stress-strain relation when the sample which had been compressed once was recompressed again.

When phase lag existed between stress and strain, processes of compression and decompression were irreversible and showed a kind of hysteresis phenomenon. The area of hysteresis loop in the process of first compression was mainly consisted of plastic deformation but in the case of recompression the area of hysteresis loop became equivalent mostly to loss energy which stemed from the viscous behavior of sample and partly to plastic deformation which had no dependency on compression time. The correlation between loss energy and other properties such as compression strain, compression time (time from the beginning of compression to the time when compression stress reached to its maximum value) and the composition of powder were investigated, and some informations were obtained, which would be described in the following.

Experimental

Apparatus——The same apparatus described in the previous paper were used. A single punched eccentric tabletting machine (Kimura, KT-2) with variable speeder and flat type punches (diameter of 16 mm) was used, to which a strain gauge and a differential transformer were connected, and signals corresponding

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[3] M. Morii, N. Takeguchi, and I. Horikoshi, *Chem. Pharm. Bull.* (Tokyo), **21**, 589 (1973).

each to stress and to strain (displacement of upper punch) were caught, amplified and used as the input of X- and Y-axis on the synchroscope. Signals from the strain gauge were not converted logarithmic scale this time, but used directly as the input. The depth of the die (or packing height; distance from the bottom punch to the upper surface of the die) was 10 mm, and the shortest punch distance, when no sample was packed in, was set beforehand to 2.68 mm.

Procedure——Sample of 550 mg was put into the die by hand, compressed at the tabletting velocity previously programmed, and recompressed at the same velocity. The relation between stress and strain in the process of compression and of decompression appeared on the synchroscope as the hysteresis loop, thenceforth the change of viscous behavior during these processes were photographed, when the composition of sample and tabletting velocity were varied.

Since X-axis has dimmension of force (compression stress) and Y-axis has dimmension of distance on the synchroscope, the area surrounded by hysteresis loop at once expresses loss energy in the process of recompression. Accordingly, accurate value of loss energy can be calculated by calibrating the relation between pressure and displacement of upper punch appeared in X-axis. The compression time, which was defined as the time length from the beginning of first compression of powder to the time when stress became maximum, wase measured by putting the signal from strain gauge onto Y-axis on the synchroscope, and the sweep signal which was synchronized to tabletting velocity, onto X-axis. And in the recompression process the authors used as a parameter the compression time which wase obtained from first compression.

Sample——Following six samples were used. Sample (A) is crystalline cellulose (PH101); (B), crystalline cellulose with 20%(w/w)of lactose; (C), with 50%(w/w)of lactose; (D), with 0.5%(w/w)magnesium stearate; (E), with 1.0%(w/w)of magnesium stearate; and (F), with 3.0%(w/w)of magnesium stearate.

When the weight of sample and the minimum punch distance were kept constant, the maximum compression stress varied between the range of 2.65×10^8—3.10×10^8 dyn/cm^2, according to the composition of sample. When the composition of sample was the same, however, the maximum value of compression stress did not change and kept almost constant, independent of the compression velocity.

Result and Discussion

An example of hysteresis loop drawn on the synchroscope in the process of recompression is illustrated in Fig. 1. In the figure, the curve $\overline{ab}$ shows the process of compression and $\overline{bcda}$ shows that of decompression. It is obvious from the figure that the maximum value of stress (point b) and the maximum value of displacement (point c) does not agree. This phenomenon is the result of the viscosity of sample, and from the area of hysteresis loop, loss energy can be calculated. In a strict sense, the area of hysteresis loop does not express only loss energy based on the viscosity of sample, but includes the plastic deformation of sample, the die friction, and the energy dissipation caused by the electrostatic force between sample and die. Here the value of plastic deformation is defined as the difference of thickness of sample before and after the recompression. The energy dissipations based on die friction and electrostatic force were so small that they were assumed to be negligible. For one of the sample the dependency of plastic deformation on compression time is illustrated in Fig. 2. The value of plastic deformation in the recompression was 0.3—0.6 mm, and the thickness of sample could be thought to be constant when the compression time was varied. So the authors assumed that the effect of plastic deformation on the total energy loss was independent of the compression time and was constant. Hence the variation of loss energy when the compression time was varied could be thought, for the most part, to be the result of the viscosity of sample, and from this experiment the authors could discuss the compression time dependency of viscous behavior.

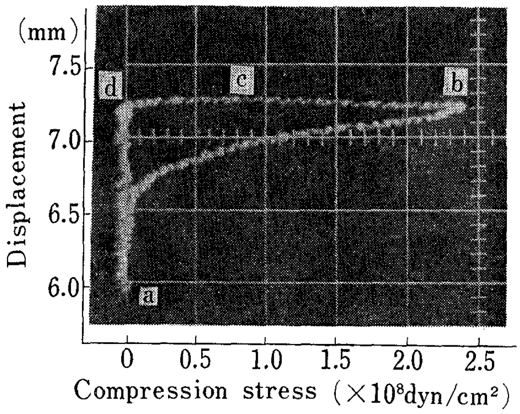

Fig. 1. Hysteresis Loop in the Process of Repeated Compression

sample: 1:1 mixture of crystalline cellulose and lactose
Compression time is 0.125 sec.

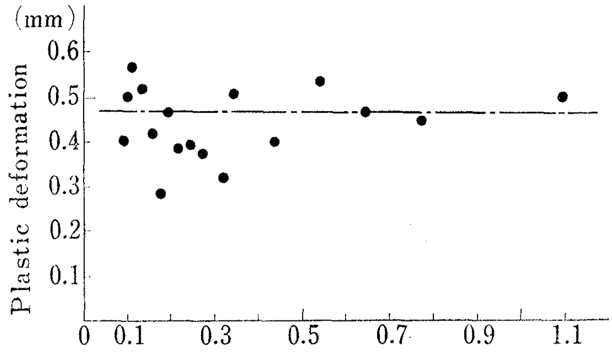

Fig. 2. Time Dependency of Plastic Deformation of Crystalline Cellulose Tablet

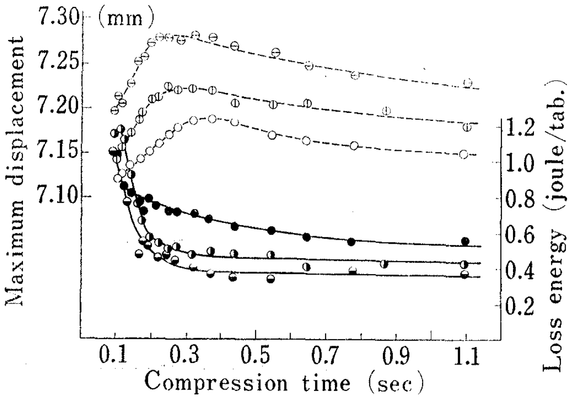

Fig. 3. Realtions between Loss Energy and Compression Time and between Maximum Displacement of Upper Punch and Compression Time

Mark	Sample	Loss energy	Maximum displacement
A	crystalline cellulose	—●—	···○···
B	4:1 mixture of crystalline cellulose and lactose	—◑—	···⦶···
C	1:1 mixture of crystalline cellulose and lactose	—◒—	···⊖···

When the dynamic compression process of powder is expressed with viscoelastic models, the viscoelastic behavior is shown to be non-linear phenomenon, and since the measurement is not carried out in a steady state but in a transient state, the coefficient of viscosity of sample can not be calculated directly from the shape of hysteresis loop and the value of loss energy. However, it is possible to determine relative values of viscous behavior and compare these values with each other. Viscous behaviors of powder in the process of dynamic compression consist of what is due to the physical character of sample itself and what is caused by the packing structure of powder, however, the latter is considered to have much greater influence on the viscous behavior. Accordingly, detail informations about viscous behavior of sample is considered to be much useful to know the change of packing structure of powder during the process of dynamic compression. Fig. 3 illustrates the case when crystalline cellulose and crystalline cellulose with lactose were used as samples, where changes of maximum displacement of the upper punch (-○-) and loss energy (-●-), against the variation of compression time were plotted. As for the maximum displacement of the upper punch, that is maximum strain, the maximum value appears in each case at a certain compression time, which means that there exist the most proper compression time (compression velocity) for tabletting. The value of loss energy, on the other hand, becomes monotonously larger as the compression time becomes shorter, and this fact shows that the influence of viscosity on compression process becomes greater as compression velocity increases. At the case when crystalline cellulose (A) is used as a sample, the value of loss energy shows the tendency of a simple increase, but when the sample of (B) or (C) is used, it keeps a constant value until compression time decreases to a certain point, and rapidly increases after the compression strain passes the maximum value. This phenomenon implies that the structural transition in which the viscous behavior of the sample varies remarkably, is taking place in the process of dynamic compression.

Changes of loss energy and of compression strain when magnesium stearate as lubricant was added to crystalline cellulose with different weight % of 0.5% (D), 1.0% (E), and 3.0% (F), are illustrated in Fig. 4. The change of loss energy corresponding to the decrease of compression time was similar to the case of crystalline cellulose only, and showed the tendency of simple increase, and there existed no remarkable difference among (D), (E) and (F). The amount of loss energy was always larger than that of crystalline cellulose. Since compression stress decreases generally when lubricant is added, it is considered that the loss energy of

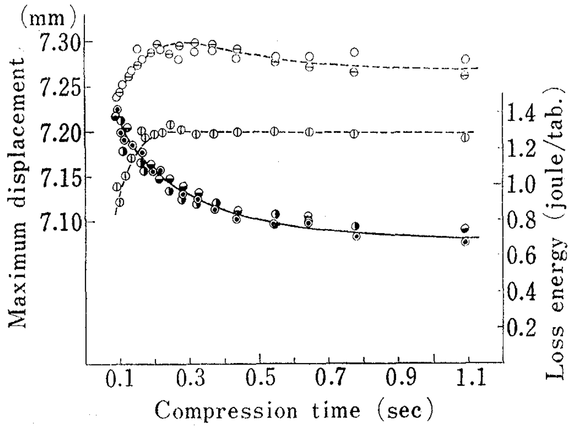

Fig. 4. Relations between Loss Energy and Compression Time and between Maximum Displacement of Upper Punch and Compression Time

Mark	Sample	Loss energy	Maximum displacement
D	crystalline cellulose with 0.5% magnesium stearate	—⊙—	···○···
E	crystalline cellulose with 1.0% magnesium stearate	—◐—	···⊖···
F	crystalline cellulose with 3.0% magnesium stearate	—◓—	···⊖···

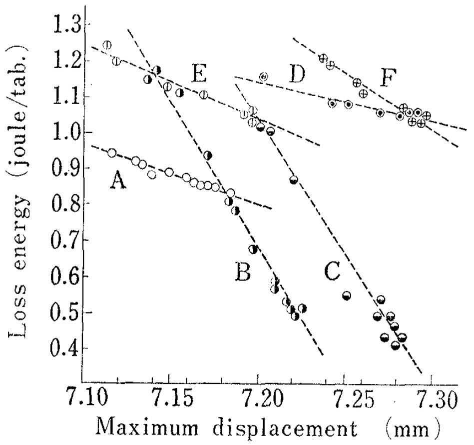

Fig. 5. The Relationship between Loss Energy and Maximum Displacement of Upper Punch after the Maximum Value of Strain

Mark A: crystalline cellulose
B: 4:1 mixture of crystalline cellulose and lactose
C: 1:1 mixture of crystalline cellulose and lactose
D: crystalline cellulose with 0.5% magnesium stearate
E: crystalline cellulose with 1.0% magnesium stearate
F: crystalline cellulose with 3.0% magnesium stearate

sample in the presence of magnesium stearate becomes larger, if these values are compared under the condition of the same compression stress, and it was clarified that increase of loss energy occurred from decrease of friction among granules of sample. The values of maximum displacement of upper punch in the case of 0.5% (D) and 3.0% (F) were greater than the value in the case of 1.0% (E). These values in the case of (D) and (F) had almost the same value and the same tendency. As for (E), it had reproducibility and it is thought to exist the minimum point around 1.0%, but its reason could not be clarified in this experiment.

Fig. 5 illustrates the relation between maximum displacement of the upper punch and loss energy after the maximum strain, that is, in the range where compression time is shorter than its maximum value. For all samples good linear relations were set up between maximum displacement and loss energy. As for samples with the presence of magnesium stearates the slopes of line were the same with that of crystalline cellulose only, whereas for the samples with lactose the slopes of line were steeper. Since maximum compression stresses were independent of compression time in this experiment, it is very reasonable to be considered that the fact that compression strain decreases after the maximum strain, is the result of increase of energy loss occurred from the viscosity of sample. Although this experiment is on the viscous behavior when sample which has been compressed once is recompressed, it is easily presumed that similar phenomenon is taking place when powder is first compressed to tablet. But the detailed behaviors of the processes of compression and of repeated compression are very complex and far from being well understood and the studies on these problems are quite few. In view of this, systematic viscoelastic analysis of repeated compression process will be of considerable interest and the authors are taking further studies.