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Evaluation of Creep Curves from the Process of Dynamic Compression[1)]

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To analyze the process of dynamic compression, especially in tabletting, a new method of drawing curves that corresponds to the normal creep curve in the static compression process was devised. The creep curve obtained by this method can be expressed using viscoelastic three elements model like $E=(P/\gamma_1)^m+(P/\gamma_2)^n\cdot[1-\exp(-t/\tau)]$, where E is compression strain, P, compressed force, t, time, τ, retardation time and γ_1, γ_2, m, n are constants. This equation can also be treated as in the case of normal creep equation.

To analyze the compression process of powder, many experiments have been carried out rheologically, and in the field of pharmaceutical engineering, there have also been a few reports[3)] on analysis of the process to make tablets using viscoelastic models. The method is mainly concerned with the analysis of stress-relaxational phenomenon that powder exhibits after its static compression, instead of the one concerning the dynamic compression while tabletting.

In order to analyze the process of dynamic compression of powder, the authors have studied the relationships among stress, strain and time while tabletting, and has been devised a new method of drawing the curve that corresponds to the creep curve in the process of static compression. The curves obtained by this method can express viscoelastic behaviors in extremely short period, and can also be measured under any pressure given. These curves can be formulated by using viscoelastic three-elements model, and the equation thus obtained can also be resolved as in the case of normal creep equation. By analyzing this creep equation, several informations about the distribution of retardation time, the most effective compression velocity and the distinction between plastic deformation and viscoelastic one were obtained.

Experimental

Apparatus——The apparatus is to draw the stress-strain curve of powder in the compression process on a synchroscope by a strain gauge and a differential transformer, and the block diagram is shown in Fig. 1.

1) Tabletting Machine: A single punched eccentric tabletting machine (Type KT-2) with a variable speeder of Ringcorn type which was set from 8 to 80 rpm, and flat type punches, diameter of 16 mm, were used. The lower punch was fixed so as the maximum pressure becomes 1.0×10^9 dyn/cm^2, and compression was done only by the motion of the upper punch.

2) Measurement of the Strain of the Compression: On a side of the upper punch was set a strain gauge which variance of electric resistance during the compression was converted to the change of voltage by a strain meter, and was used as the input of X-axis in the synchroscope through the logarithmic amplifier. The calibration of the strain gauge was carried out by setting off the upper punch with the gauge from the tabletting machine, and by compressing the upper punch with an oil press under static condition.

3) Measurement of Punch Distance: In order to measure the punch distance, a differential transformer was set between where the upper and the lower punch had been fixed to the tabletting machine. The punch distance was converted to electric voltage by a displacement meter and was used as the input of Y-axis in the synchroscope. When the differential transformer was set as mentioned above, no correction of mechanical rattles was needed. The strain of the punch itself was found to be negligible.

1) Location: *3190 Gofuku, Toyama.*
2) Presented at the 91th Annual Meeting of the Pharmaceutical Society of Japan at Fukuoka, Apr. 1971.
3) T. Anmo, M. Washitake, T. Kurashige, and M. Naritomi, *Yakuzaigaku*, **26**, 267 (1966); T. Anmo, M. Washitake, T. Kurashige, M. Naritomo, H. Ono, and M. Asano, *Yakugaku Zasshi*, **88**, 859 (1968); T. Anmo, M. Naritomi, H. Ono, and M. Asano, *ibid.*, **89**, 359 (1969).

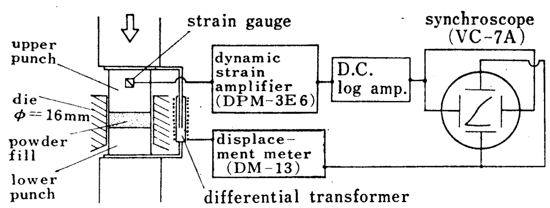


Fig. 1. Block Diagram of Apparatus

4) Photographing of Stress-strain Diagram: Electric signals from the strain gauge and the differential transformer were entered in as X- and Y-axis of the synchroscope, and the trace of the compression process was drawn on its Braun tube, which was photographed. Carrier waves of the strain gauge and the transformer were 5.0 and 1.2 kHz which were high enough, and the frequency characteristic of the synchroscope were also good enough to express the trace that had few phase distortion even in high compression velocity.

Material——Crystalline cellulose (PH101) was kept in a desiccator containing calcium chloride, for a week. Average diameter of particles was 37.6 μ, and the standard deviation, 13.5 μ.

Procedure——To fill the die, the depth of which was 11.0 mm, 550 mg of crystalline cellulose was needed. The position of the upper punch was controlled so as the value of the maximum stress to become 1.0×10^9 dyn/cm^2 when compressed. The velocity of the compression was varied by ten grades. To draw the curve of the punch distance against compression time, the synchroscope was regulated by a trigger level control circuit so that the moment when the upper punch had touched the sample in the die, the trace appeared on the Braun tube, which was photographed.

Result and Discussion

I) Stress-strain Diagram of the Compression Process

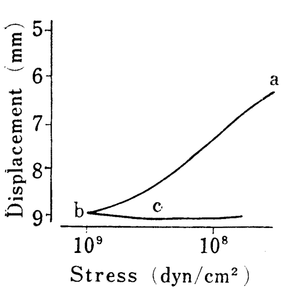
The relationship between the stress and the punch distance in the compression process is shown in Fig. 2-1. When compression strain (E) is defined as the ratio of initial depth (L_0) of die filled with sample and punch distance (L) in the compression process, Fig. 2-1 can be regarded as the stress-strain diagram. In Fig. 2-1 (a) is the point where the trace begins, (b), where the pressure becomes maximum, and (c), where the strain becomes maximum. As the horizontal axis is graduated to the logarithmic scale, the curve from (a) to (b) should be straight, if the process of compression follows Bal'shin's equation.[4] However in this experiment, using crystalline cellulose, it did not draw a straight line. Pressure needed to produce a certain deformation increases with compression velocity, which is attributable to the viscosity of sample. Generally in the dynamic compression of viscoelastic materials, the phase of strain laggs behind that of stress[5], and actually in this experiment the phase lag was observed; even when stress passed point (b), which was maximum value, and began to decrease, the strain continued to increase until point (c).

Fig. 2-2 shows curves of punch distance versus time. The trace (1) in Fig. 2-2 is the case when tabletted without sample and (2) is one with sample. In each case, at the maximum point of the displacement appears a plateau in the curve, which shows that the motion of the upper punch draws no sinusoidal curve because of the mechanical rattles.

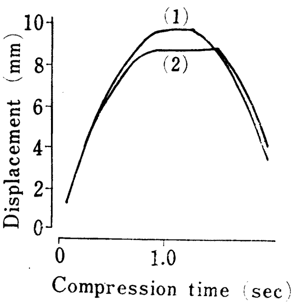
II) Creep Curve of Crystalline Cellulose

The stress-strain curves such as in Fig. 2-1 were photographed by changing the compression velocity as many times as possible, and from each curve the strain under a constant value of stress was obtained. A group of curves corresponding to each stress can be obtained, when these strains are plotted against the compression time which is measured from Fig. 2-2. The results are shown in Fig. 3. These curves show the behavior of strain versus time when samples are compressed in a constant pressure, and are thought to be a kind of creep curve. However, since these curves are estimated from the dynamic compression process, they can not be regarded as the same with those obtained in the static compression process. The maximum values of strain in the stress-strain curves were constant, regardless of compression veloci-

4) W.D. Jones, "Fundamental Principles of Powder Metallurgy," Edward Arnold Ltd., London 1960, p. 232.
5) T. Nakagawa and H. Kanbe, "Rheology," Misuzu Shobo, Tokyo 1959, p. 535.



(Fig. 2.1)



(Fig. 2.2)

Fig. 2-1. Relationship between Displacement and Stress

whole compression time is 1.13 sec.

Fig. 2-2. Relationship betweeen Displacement and Compression Time.

trace (1): without sample
trace (2): with 550 mg of crystalline cellulose

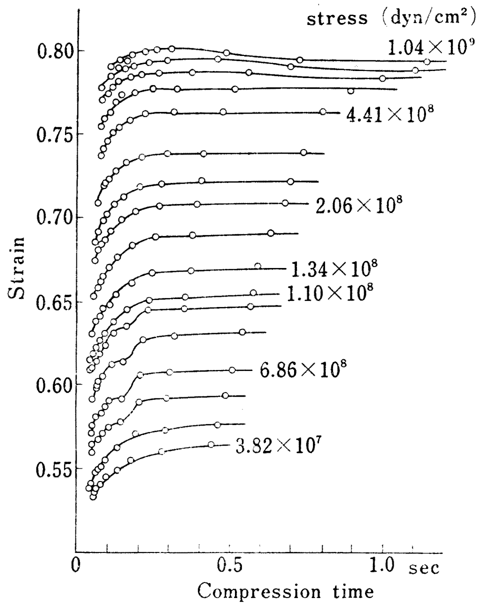


Fig. 3. Creep Curves evaluated from Dynamic Compression Process

ty, and were 1.04×10^9dyn/cm^2 in this experiment. If the maximum stress was altered, however, the creep curves obtained were not in accord with ones in Fig. 3. This phenomenon results from the sinusoidal motion of the upper punch, and because the stress of the compression is controlled with the change of the position of the upper punch, differential compression velocity of the upper punch at a certain moment differs, even in the same compression stress and in the same compression time, and consequently the difference in strain appears. Deformation process of compressed powder belongs to that of nonlinear type and is mostly the plastic deformation, and although it can not be expressed by the simple equation of creep curve, can be formulated in a similar way to the case of linear type. In regard to the creep curves in Fig. 3, when the value where (E) reached to equilibrium is named ($E_{eq.}$) and when $\log[E_{eq.}-E]$ is plotted against time, there exists a linear relation shown in Fig. 4, from which the creep curves can be expressed by a similar equation to that of the linear viscoelastic three elements model as

$$E=(P/\gamma_1)^m+(P/\gamma_2)^n\cdot[1-\exp(-t/\tau)] \qquad (1)$$

where P is the stress of compression, γ_1 and m are constants regarding instantaneous deformation, γ_2 and n are constants regarding retarded deformation, and τ is the retardation time.

When the creep curves in Fig. 3 are applied to this equation, a good agreement is obtained in the middle region of stress ($4.0-1.1\times10^8$ dyn/cm^2), but in the upper and the lower region, no agreement. That is, at the maximum compression stress (1.04×10^9 dyn/cm^2) and in its neighbor, the maximum peaks appear in the creep curves. The existence of those maximum values suggests that there exists the most effective compression velocity for tabletting.

In the low stress region, from 1.1×10^8 to 4.0×10^7 dyn/cm^2, the experimental values can not be written in the form of the creep equation which has single retardation time; there may be a certain distribution in retardation time. It may be because that within those region of stress, the packing structure of powder is influenced by compression velocity, and becomes unstable. On the other hand, in the stress region lower than 4.0×10^7 dyn/cm^2, experimental values again can be expressed with a curve of a single retardation time, which means that powder maintains a stable packing structure.

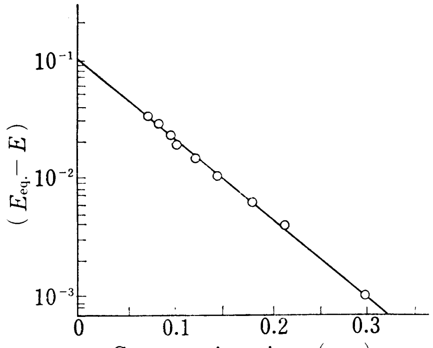


Fig. 4. Relation between [($E_{eq.}$)-(E)] and Compression Time.
stress: 3.14×10^8dyn/cm^2

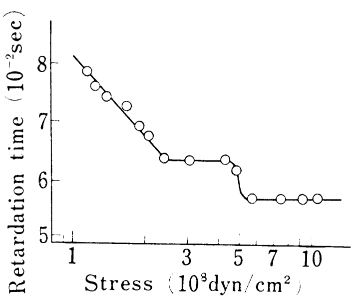


Fig. 5. Relationship between Retardation Time and Stress

The relation between τ and P are illustrated in Fig. 5. Apparently from the figure, the retardation time is divided into three parts according to the value of stress. One is where the stress is under 2.0×10^8 dyn/cm^2, where τ becomes smaller as the stress increases. The others are within each region from 2.0×10^8 to 5.0×10^8 dyn/cm^2 and from 5.0×10^8 to 1.04×10^9 dyn/cm^2, where values of τ keep constant, and from this fact it can be said that the packing behavior of powder does not change and the processes of creeping are identical. The values of τ in this experiment are all smaller than 0.1 sec, and creeping is accomplished in a short time.

The results shown in Fig. 6 were obtained, when $E_{eq.}$(equilibrium strain), $E_{inst.}$(instantaneous strain) which was obtained when the line in Fig. 4 had been extrapolated to time zero, and $E_{ret.}$(retardation strain) which was obtained by introducing $E_{eq.}$ and $E_{inst.}$ into the equation (1), were plotted against the stress on homologarithmic graph. In Fig. 6, the white dots (-○-) show the values of $E_{eq.}$, the half white dots (-◑-), the experimental values of E when $t=0.104$ sec, and the dotted line shows the extrapolated value of $E_{inst.}$ and the broken line, the calculated value of $E_{ret.}$ As in the case of compression of powder or granules, $E_{inst.}$ is mostly the plastic deformation, $(p/\gamma_1)^m$ in the equation (1) is nearly equal to the value of plastic deformation, and $E_{ret.}$ is nearly equal to the one of viscoelastic deformation $(P/\gamma_2)^n$. Consequently $E_{eq.}$ is considered to be the summation of these two deformations, and its experi-

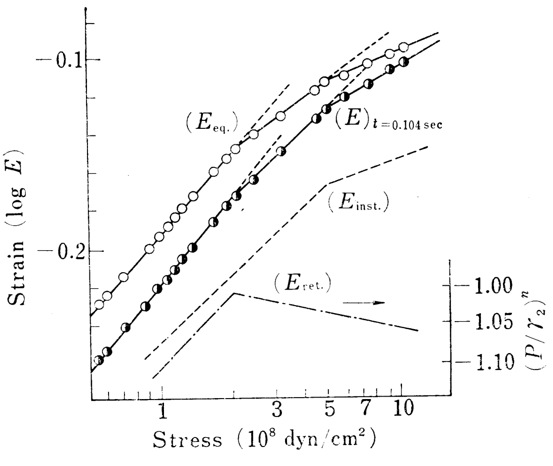


Fig. 6. Relationship between Stress and Strain

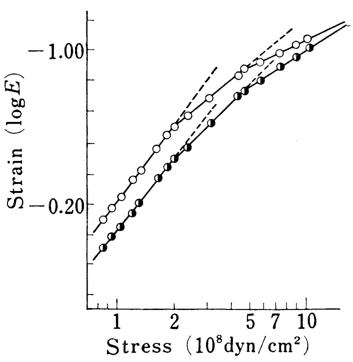


Fig. 7. Relation between Stress and Strain.
—○—: whole compression time=0.310 sec
—◑—: whole compression time=0.107 sec

mental values are expressed by two lines, each of which had two bending points at 2.0 and 5.0×10^8 dyn/cm^2. This means that there are stepwise changes in the packing structure of crystalline cellulose, and from the fact that each of the two bending points of $E_{eq.}$ corresponds to $E_{inst.}$ and $E_{ret.}$, the bending at 2.0×10^8 dyn/cm^2 is considered to be based on the viscoelastic character of powder, and one at 5.0×10^8 dyn/cm^2 on the plastic character. Thus changes of packing structure are able to discriminate into two parts, the plastic and viscoelastic behaviors. Besides, when the compression process in Fig. 2-1 is plotted on a logarithmic graph, it is expressed as a line shown in Fig. 7, which bends at the same value of stress to that in Fig. 6, and it is also affirmed that the stress by which the structural change occurs is constant, regardless of tabletting velocity.

Conclusion

The creep curves obtained by the method mentioned above, can be formulated by the same way to that of linear viscoelastic phenomenon, and this method was proved to be quite effective to analyze the dynamic compression process which had not been investigated. When crystalline cellulose is used as the sample, a maximum value appears in the creep curve, which suggests that the pattern of packing structure changes by compression velocity, and that compression is not so effective under low compression velocity, although it is generally considered to be easy to do. By the method described in this report, the most suitable compression velocity can be found. In addition, by analyzing these creep curves, it is possible to clarify how far the change of the packing structure during dynamic compression attributes to the plastic deformation and to the viscoelastic deformation.