Bonding Mechanisms and Hysteresis Areas in Compression Cycle Plots

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Abstract □ It was shown previously that if compression of a powder is treated in an analogous manner to the compression of a nonporous solid, a plot of the transmitted pressure versus the applied pressure should have a hysteresis area that is linear with respect to the maximally applied pressure if plastic deformation occurs. However, if brittle fracture occurs, the area should be quadratically related to the maximally applied pressure. A series of compounds expected to yield by plastic deformation was tested, and they were found to give the quadratic relationship. Thus, it is indicated experimentally that the assumptions made in general in treating compression cycles may be partially invalid.

Keyphrases □ Powders—compression cycles at various maximum pressures for substances expected to undergo plastic deformation □ Tablet manufacture—compression cycles at various maximum pressures for substances expected to undergo plastic deformation □ Compression cycles—compression at various maximum pressures of substances expected to undergo plastic deformation

In tablet manufacture, a powder or granulation is placed within a die and then compressed between two punches. The process can be considered as being composed of five stages (1):

1. The particles rearrange so that the powder attains its closest packing.

2. The particles then deform elastically.

3. At stresses or pressures above the elastic limits, the particles either deform plastically or fracture. Either process is assumed to lead to bonding between the particles and, therefore, to tablet formation (2).

4. The upper punch is released, and the stresses relax.

5. The tablet is ejected.

If the die wall pressure (F) or lower punch pressure (P') in Stages 1–4 is monitored as a function of the upper punch pressure (P), then a compression cycle results.

BACKGROUND

Long (3) and Leigh et al. (4) analyzed the theoretical relationships $P' = f(P)$ and $F = f(P)$ and arrived at relationships that have been useful in investigating the physics of powder compression. Carstensen (5) and Carstensen and Toure (6) applied the suggested model, which involves an analogy between the behavior of a nonporous mass and the powder in question, to treatment of the areas of the hysteresis loops described by the functions $P' = f(P)$ and $F = f(P)$. They cautioned that the assumptions used might be unrealistic in some aspects and concluded that if the assumptions held, then bonding by plastic deformation should lead to a linear relationship between the hysteresis area (A) and the maximally applied pressure (P_m):

$$A = a'P_m + b' \tag{Eq. 1}$$

If brittle fracture occurred, then the relationship would be:

$$A = c + bP_m + aP_m^2 \tag{Eq. 2}$$

or if c and b were small, it would be simply:

$$\ln A = 2 \ln P_m + \ln a \tag{Eq. 3}$$

where a, b, and c are constants. One interesting note in the few available

Table I—Regression Values for Cycle Areas (A) as a Function of Maximum Pressure (P^*) According to Eq. 4

Formula	Correlation Coefficient	α	β
Polyvinyl chloride	0.998	1.90	−1.66
Polyvinyl chloride and excipients[a]	0.998	1.82	−0.79
Polyvinyl alcohol	0.996	1.94	−1.67
Polyvinyl alcohol and excipients[a]	0.998	2.2	−3.0
Polyvinyl alcohol–polyvinyl chloride copolymer	0.983	2.13	−2.83
Polyvinyl alcohol–polyvinyl chloride copolymer (50%) and excipients[a]	0.998	1.94	−1.68
Polyvinyl alcohol–polyvinyl chloride copolymer (60%) and excipients[a]	0.999	2.05	−2.24
Polyvinyl alcohol–polyvinyl chloride copolymer (70%) and excipients[a]	0.998	2.15	−2.81
Polyvinyl alcohol–polyvinyl chloride copolymer (80%) and excipients[a]	0.999	1.97	−1.99

[a] The excipients were as stated in the text; the granulation liquid was isopropanol, except in the last case where methylene chloride was used.

cases is that sodium chloride appears to bond by brittle fracture rather than by plastic deformation, as has been generally assumed.

The aims of the present study were to test the compression cycles at various maximum pressures for substances that are expected to deform plastically beyond the elastic limit and to test which equation (Eq. 1 or 2) is valid.

EXPERIMENTAL

The compositions of the tablets are shown in Table I. In three cases, the polymers (polyvinyl alcohol[1], polyvinyl chloride[2], and a copolymer of the two compounds[3]) were compressed as they were. The particle sizes of the polymers were 250–400, <5, and 80–125 μm, respectively. In the remaining cases, granulations of the polymers were used consisting of 20% diphenhydramine hydrochloride, 1.5% povidone, 50% polymer (unless stated otherwise) and an amount of tribasic calcium phosphate (q.s.). With the copolymer, a series of compositions was compressed with increased polymer content (60 and 70%).

The granulations were prepared by mixing the powders without povidone and then granulating them with a 3.75% solution of povidone in either isopropanol or methylene chloride. The granulation was dried at 60° for 15 min in a small fluid bed drier[4]. Then the granulation was passed through an oscillating granulator with a 1-mm screen opening, after which the 0.2–0.8-mm fraction was separated out by screening and used for compression.

Compression was carried out on a single-punch tablet machine[5] equipped with strain gauges and operated at a speed that gave a cycle time of 1/60 sec; 0.5 g of powder or granulation was used for the compression, and 12-mm flat-faced punches were employed. Data were reduced to sets of corresponding P and P' values, and a hysteresis plot (Fig. 1) was made for each. Areas of the hysteresis loops were determined with a planimeter.

RESULTS AND DISCUSSION

A typical set of compression cycles (with three different maximal

[1] Rhodopas, H, Rhone-Poulenc, France.
[2] Pevikon, 637P, Seppic, Paris, France.
[3] 15 PVA85PVC, Solvic, Solvay, France.
[4] Glatt, Basel, Switzerland.
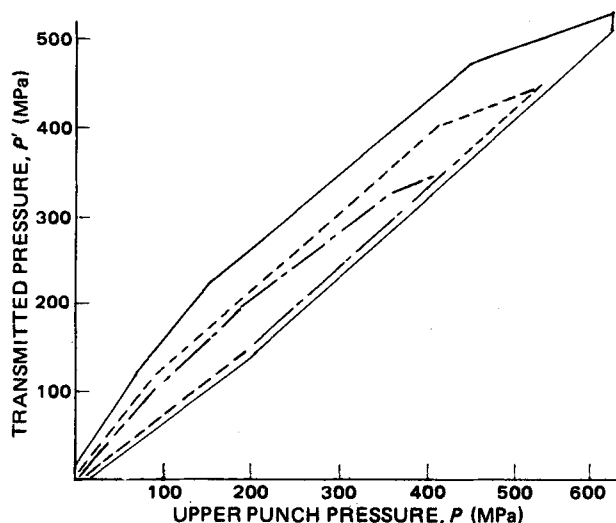[5] Frogeais type AO, Societe Edmond Frogerais, Vitry-sur-Seine, France.

Figure 1—*Pressure cycles at three maximal pressures for polyvinyl alcohol–polyvinyl chloride copolymer (60%) and excipients. Two other cycles at other maximal pressures are not shown for pictorial clarity.*

pressures) is shown for one formulation (copolymer and excipients) in Fig. 1. Five maximal pressures were employed in each case, but only three are shown for pictorial clarity. Each point of an individual cycle is the mean of six experimental determinations, so each cycle is an average cycle; the standard deviation of a sample of six points was ~4%. In all cases, the areas (A) of the hysteresis loops were plotted *versus* the maximal pressure, P^*, resulting in relations of the type:
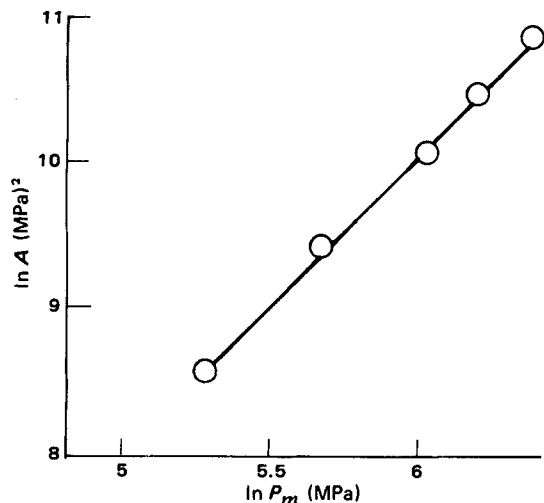

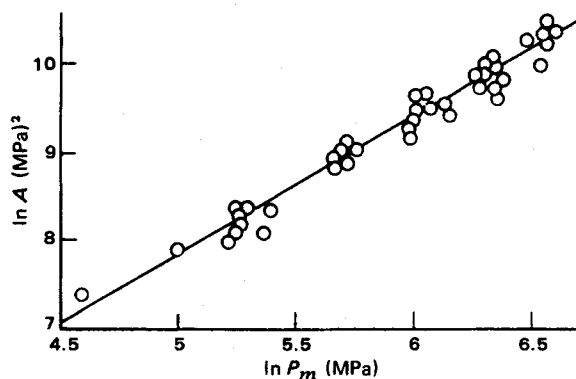**Figure 2**—*Areas of the hysteresis loops of Fig. 1 plotted according to Eq. 4.*

Figure 3—*All of the experimental hysteresis loop areas plotted according to Eq. 4.*

$$\ln A = \alpha \ln P_m + \beta \qquad \text{(Eq. 4)}$$

The data from the formulation in Fig. 1 are plotted in this fashion in Fig. 2, and the linearity is obvious. The slopes and intercepts obtained by least-squares fitting are shown in Table I. All of the slopes are close to 2. The fact that the plots are similar for all of the compositions is not surprising since the polymers can be expected to exhibit comparable behavior due to their chemical similarity. The fact that the data are comparable allowed graphical presentation of all of the experimental (area) data on one plot (Fig. 3).

The surprising feature is that the quadratic relationship:

$$A = aP_{\text{max}}^2 \qquad \text{(Eq. 5)}$$

occurs, as predicted for brittle fracture. Polymers intuitively should give rise to plastic deformation, and Eq. 1 (the linear relationship) should hold rather than Eq. 2 (or Eq. 3). Carstensen and Toure (6) pointed out that some assumptions made in conventional modeling of compression cycles may be unrealistic. The solid is highly porous at the onset of the cycle and slightly porous to nonporous (practically in some cases) at the end but, by analogy, is compared to a solid, nonporous body at all stages. Equations 1 and 2, therefore, are no more valid than the assumptions made; and since the polymers used here probably do not bond by brittle fracture, it may be assumed that one or more of the assumptions made in deriving the conventional cycle equations (whether for the individual line segments or for the areas) are unrealistic.

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