(2) R. M. Jacobs, G. L. Regniér, and C. Crisan, U. S. pat. 2,944,061(July 5, 1960) (Societe des usines chemiques Rhone-Poulenc).

(3) C. Cosar and L. Jalu, *Ann. Inst. Pasteur*, **96**, 238(1959).

(4) L. Sylvestre and Z. Gallai, Union Med. Canada, **89**, 334 (1960); through *Chem. Abstr.*, **54**, 18789*a*(1961).

(5) N. R. Boazaine and J. M. Desranleau, *ibid.*, **89**, 759(1960); through *Chem. Abstr.*, **54**, 18789*a*(1961).

(6) M. Back, *Arzneimittel-Forsch.*, **11**, 587(1961).

(7) P. O. Kane, *J. Polarog. Soc.*, **7**, 58(1961).

(8) A. Danek, *Dissertationes Pharm.*, **13**, 107(1961); through *Anal. Abstr.*, 3909(1961).

(9) C. Cosar, M. Dubost, P. Dubost, P. Devoize, and M. Palliere, *Ann. Pharm. Franc.*, **20**, 872(1962).

(10) L. Vignoli, B. Cristau, F. Gouezo, and C. Fabre, *Chim. Anal.* (*Paris*), **45**, 499(1963).

(11) J. Breinlich, Deut. Apotheker-Z, **104**, 535(1964).

(12) Private communication, Searle, Inc., Chicago, Ill.

ACKNOWLEDGMENTS AND ADDRESSES

Received May 7, 1969 from *Temple University School of Pharmacy, Philadelphia, PA 19140*

Accepted for publication July 9, 1969.

Presented to the Pharmaceutical Analysis and Control Section, APhA Academy of Pharmaceutical Sciences, Montreal meeting, May 1969.

The material examined was donated by Searle, Inc., Chicago, Ill.

* Present address: Zavod Za Ispitivanje i Kontrolu Lijekova, SRH. Mlinarska cesta 38, Zagreb, Yugoslavia.

TECHNICAL ARTICLES

Shear Cell Measurements of Powders: Proposed Procedures for Elucidating the Mechanistic Behavior of Powder Beds in Shear

E. N. HIESTAND and C. J. WILCOX

Abstract ☐ A variety of procedures for the use of a shear cell are devised in hopes of elucidating mechanistically the behavior of powders in shear. The processes considered are plastic deformation at regions of true contact and structural changes in the powder bed. Possible structure changes are: (*a*) consolidation or dilation; (*b*) blockage to resist the continuation of motion in the same direction; and (*c*) particle orientation. A series of pulls to just initiate shear is used. These are made either monodirectionally, *i.e.*, all pulls in the same direction, or bidirectionally, *i.e.*, consecutive pull directions reversed. A series of pulls proceeds until a steady state or plateau condition exists. A relative shear force, M, is defined as $M = (\mu_1 - \mu_p)/\mu_p$, where μ_1 and μ_p are the respective friction coefficients using first pull and plateau data. The retention of plateau-condition-shear-strength upon removal of the applied load is obtained by measuring the shear force for the first pull after reducing the load. An index of retention value is defined. Also, an indication of the relative extent of conditioning at plateau condition for various loads is based on a comparison of the shear force observed for one additional pull after increasing the load to an arbitrary constant value.

Keyphrases ☐ Powders—shear cell measurements ☐ Shear cell—procedures for use ☐ IP patterns ☐ Forces shear—reduced and increased load ☐ Structural changes—powder bed

In this communication a series of procedures will be proposed, then the experimental use of these will be described. The objective is to develop a mechanistic interpretation of the resistance to shear of powder beds. New parameters and indices will be defined when needed for simplification of the discussion.

The procedures are based on variations of shear cell studies reported previously (1). The primary reference values are the friction data in the form of a plot of the shear force, τ, *versus* the applied load, mg. The simple friction law, Eq. 1, adequately describes these friction data.

$$\tau = \mu(mg + h) \qquad \text{(Eq. 1)}$$

where τ = force required to initiate shear, μ = friction coefficient, mg = load normal to the shear plane, and h = "apparent" cohesion.

As stated in the earlier publication, the observed values of μ and h depend somewhat upon cell design, operational procedure, and the history of the powder bed. Reproducible values are obtained readily when a specific cell and a standard procedure are adopted. The values obtained are useful for making comparisons or detecting changes in the properties of a powder. However, μ and h alone are not adequate to indicate many of the characteristics of a powder bed. Additional meaningful properties must be identified and measured.

Occasional reference will be made to the properties that are associated with good flowability. It is recognized that flowability is not a uniquely defined property. It has meaning only in reference to the conditions under which flow is to occur. Nevertheless, no matter how elusive its precise definition may be, common usage has established a qualitative, intuitive understanding of its meaning. Hereinafter flowability is used in this qualitative connotation.

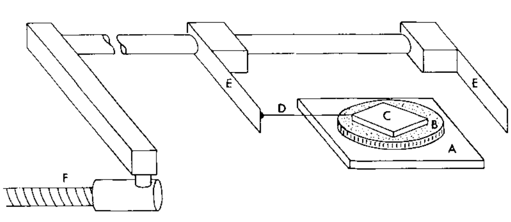

Figure 1—*Schematic arrangement of apparatus. Key:* A, *bottom plate;* B, *powder bed;* C, *top plate;* D, *tow line;* E, *strain gauges (both are connected when bidirectional pulling is used);* F, *jack.*

PROPOSED PROCEDURES

Apparatus—The shear cells[1] for this work have been described previously (1). Only a few relevant aspects will be repeated here. Figure 1 shows the cell arrangement diagrammatically. The powder is located between two solid surfaces, analogous to a sandwich filling. Two different sandwiches are shown in Fig. 2—*viz.;* (*a*) the sandpaper-powder-sandpaper (SPS cell) and (*b*) the tablet-powder-tablet (TPT cell). In the figure the top plate is inverted to show its surface texture.

Experimental Procedure—All of the data reported in this article have been obtained with the shear cell. However, numerous variations of procedure are used. To facilitate the discussion of these data, a somewhat complex set of symbols has been developed. The symbols were chosen to be both concise and mnemonic. Often the experimental procedure for obtaining the data is coded into the symbol. The complete meaning of the symbols are given in the text as they are introduced. For the reader's convenience, a table of nomenclature is included at the end of the text. In some instances, the orderly evolvement of the symbol is explained for mnemonic purposes.

Most of the proposed procedure is the same as reported previously. The shear force is observed for a sequence of pulls, each of which just initiates shearing of the powder bed.[2] Eventually a constant value of the shear force is obtained for each sequential pull. This shear force is called the plateau value, τ_p. The symbols $\tau_p c$ will be used to indicate the plateau condition shear force observed when the applied load is equal to c.

IP Patterns—The recorder chart record of the shear forces for the sequence of pulls between the initial and plateau condition is called the IP pattern. The three general types of IP patterns observed (1) are shown in Fig. 3. I-IP designates increasing shear forces for the sequence of pulls, D-IP refers to decreasing forces, and P-IP indicates that a peak value of shear force occurs between the initial and plateau condition of the bed.

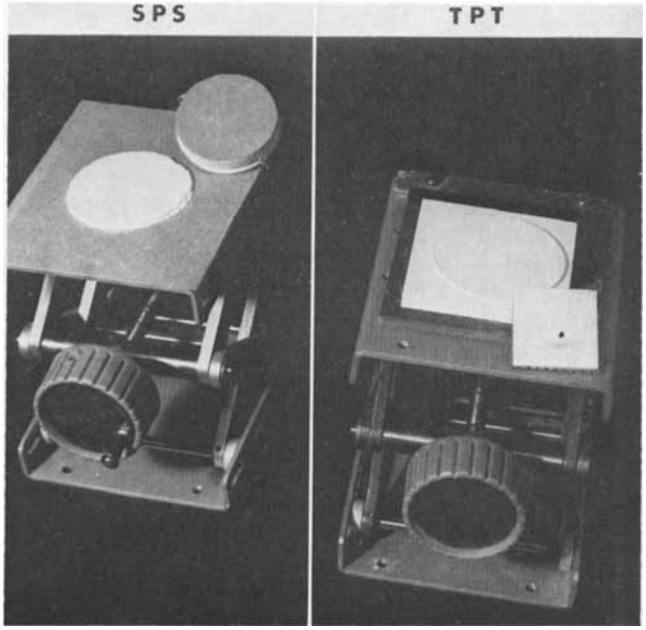

Figure 2—*Two designs of shear cells used:* SPS, *sandpaper-powder-sandpaper;* TPT, *tablet-powder-tablet.*

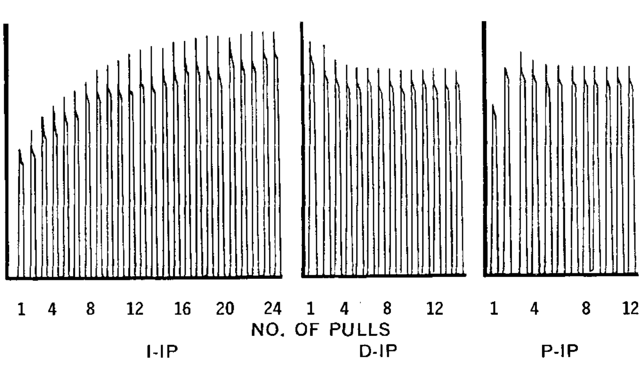

Figure 3—*Recorder chart patterns produced in the sequence of pulls when going from the initial to plateau condition of the bed. Key:* I-IP, *increasing shear strength from initial to plateau;* D-IP, *decreasing shear strength from initial to plateau;* P-IP, *peak shear strength between initial and plateau.*

The sequences of pulls are made in two different ways—*viz.;* (*a*) all pulls in the same direction (monodirectional pulling); and (*b*) each consecutive pull is made in the opposite direction to alternate the direction of pull (bidirectional pulling). When using bidirectional pulling with certain powders, the force at which shear is initiated may not be identifiable; but usually bidirectional data may be obtained. Bidirectional pulling requires care in alignment of the top cell element between the two strain gauges so that a twisting or rocking[3] motion is not induced.

Usually, but not always, the IP patterns are of the same type for both mono- and bidirectional pulling. The respective plateau shear force values are identified as τ_{p1d} and τ_{p2d}.

Reduced Load Shear Forces—Measurements of the shear force determined by first obtaining a plateau condition with some relatively large applied load, then removing some or all of the applied load, and finally pulling one additional time to produce shear are referred to as reduced load shear values, τ_r. Both mono- and bidirectional plateau conditions may be used to obtain τ_{r1d} and τ_{r2d} values, respectively. The only τ_{r2d} values considered herein are those obtained by making the direction of the reduced load pull opposite to that of the last pull used to obtain the plateau condition bidirectionally.

Three different procedures for obtaining reduced load values may be followed. These are:

1. The applied load is reduced to various values always from the same plateau condition obtained at a selected, constant value of the applied load. $c\tau_r x$ will be the generalized designation, *e.g.*, $200\tau_r x$ designates a series of values obtained by reducing the load by different amounts from the plateau condition obtained using a 200-g. total applied load.[4]

2. The applied load is reduced by a constant amount from various plateau conditions obtained with different applied loads, $x\tau_{cr}$.

3. The applied load is reduced to a constant value from various plateau conditions obtained with different applied loads, $x\tau_r c$.

All of these provide an indication of the degree of retention after load reduction of properties produced by shear at the larger applied load. The last of these is considered by the authors to be the most useful for the purpose of this communication. Based on it, a method of assigning numerical values to the retention of shear strength induced at larger loads (designated by index of retention), will be described in a later section. The first two procedures are useful to

---

[1] Three cells were described. The selection of the preferred one is based on experience. Usually the SPS cell is satisfactory.

[2] Dynamic friction coefficients could be determined, also, but only static yield forces are considered in this discussion.

[3] The cell design places the tow line slightly above the plane of shear. Intuitively, this seems undesirable; but the authors have observed that when the top plate tilts after repeated pulling in the same direction, the leading edge of the plate always is higher than the trailing edge. Lowering the tow line to the shear plane would increase the tendency to tilt in this direction. Therefore, the tow line was not lowered to the plane of shear.

[4] These measurements are not identical to but correspond in principle to the "yield loci" curves obtained with the Jenike shear cell (2).

provide additional insight into the retention under various loads. They are mentioned here for completeness and one example is given in the experimental section.

Increased Load Shear Forces—The IP patterns arise from changes in shear strength produced by shearing the bed, *i.e.*, shear-induced conditioning occurs. However, these patterns do not indicate whether the plateau conditions at all loads are beds of identical condition. An indication of relative effect of conditioning may be obtained by first attaining a plateau condition at some load, x, then increasing the load to c and making one additional pull. This last shear force, $x\tau_I c$, may be compared with $\tau_1 c$ (see *Footnote* 5) and $\tau_p c$. If $x\tau_I c$ equaled $\tau_1 c$, it would appear that no conditioning effect on the shear force occurred as a result of reaching plateau conditions with load x. If $x\tau_I c$ equalled $\tau_p c$, it would appear that plateau conditions with load x was identical to plateau conditions with load c.

A convenient reference value for c is the largest load used in the study. For example, if 1000 g. is the largest load used, then by determining $x\tau_I 1000$ for several values of x and by plotting the $x\tau_I 1000$ values *versus* the x values, a comparison with $\tau_1 1000$ and $\tau_p 1000$ may be made. These data can be plotted on the same graph as the friction data.

Both mono- and bidirectional values are useful. The bidirectional values used are those obtained by pulling after increasing the applied load in the direction opposite to the last pull direction used when attaining the plateau condition with the lesser load.

Obviously many variations of these increased load studies are possible and other methods of plotting could be used. However, the authors have chosen the above-described method because of its simplicity and ease of interpretation.

Numerical Representation of Data—*IP Patterns*—The three types of IP patterns provide a qualitative classification of powder bed properties. A more quantitative representation can be obtained from these by application of Eq. 2.

$$M = \frac{\mu_1 - \mu_p}{\mu_p} \qquad \text{(Eq. 2)}$$

or its equivalent

$$M = \frac{(\tau_1 - \mu_1 h_1) - (\tau_p - \mu_p h_p)}{\tau_p - \mu_p h_p}$$

where M is called the relative IP value, μ_1 is the friction coefficient based on first pull shear forces $\tau_1 x$, μ_p is the friction coefficient using plateau shear forces $\tau_p x$, and h_1 and h_p are the respective apparent cohesion forces.

The IP value, M, compares the initial to plateau friction coefficients so that M is positive for materials of good flowability, *i.e.*, for Type D-IP; is negative for those of poorer flowability, *i.e.*, for Type I-IP; and may be either negative or positive for Type P-IP patterns. These numbers are useful indicators but of course the precision is limited by the ability of the investigator to form the powder bed in a reproducible manner. Failure to do so will produce variations in μ_1 but has no significant effect on μ_p.

When considering the P-IP patterns, an indication of the peak shear force can be given by using an M' value defined in Eq. 3.

$$M' = \frac{\mu' - \mu_p}{\mu_p} \qquad \text{(Eq. 3)}$$

where μ' is the friction coefficient obtained by plotting peak values of the shear force.

Index of Retention—Values of the reduced load shear force obtained by Method 3 may be plotted against the τ_p value at the load from which the reduction was made, *i.e.*, $x\tau_r c$ *versus* $\tau_p x$. If no effects of the larger load are retained, the $x\tau_r c$ values are all equal and such a plot would have a slope of zero. If all the $x\tau_r c$ values are equal to the $\tau_p x$ values, the corresponding slope would be unity. This would correspond to complete retention of shear forces produced by the larger load. The authors have chosen to refer to the slope of the plot of $x\tau_r c$ *versus* $\tau_p x$ with c = (weight of disk or top tablet) as the index of retention. Because the plot is not linear, the index of retention is not a single value. Usually the largest values are observed for small reduction of load and the magnitude of these values correlate at least qualitatively with poor flowability.

Mechanistic Processes of Shear in Powder Beds—Before discussing real cases, it will be useful to list in one place a number of mechanisms that have been considered to influence the shear force. The following is not claimed to be an exhaustive list but is believed to include the most important considerations:

1. Structural Changes in the Powder Bed—(*a*) Consolidation or dilation that alters the average area of true contact between particles in the shear plane; (*b*) Mechanical blockage or pile-up that resists continuation of motion in the same direction; (*c*) Particle orientation and alignment that lead to differences between shear forces when motion is continued in a given direction and when the direction is reversed.

2. Changes in the Area of True Contact between Particles Resulting from Plastic Deformation of Particles at Contact Regions. If only one of these mechanisms were occurring in a given powder bed, the interpretation of the data would be relatively simple. It is instructive to consider these simple cases in which only a single process is occurring.[6]

Case 1(a)—Consolidation induced by load and shear would lead to an I-IP pattern with no differences between the mono- and bidirectional shear forces. The corresponding M value would indicate the relative change of friction coefficient for this consolidation. Increased load studies would indicate the relative differences in extent of consolidation with various applied loads. The index of retention would relate to the force necessary to return the powder bed to the more expanded condition characteristic of the very small load.

A powder bed that dilates each time it is sheared would not exhibit shear conditioning. Therefore, M would equal zero. The index of retention would be zero, also; and the increased load studies would yield a value of one for the relative conditioning.

Case 1(b) differs from 1(*a*) in that the monodirectional shear forces will be larger than the bidirectional forces. The monodirectional patterns would have a negative M value and the bidirectional a zero value. The index of retention would be zero for bidirectional pulling but could have a large value for monodirectional pulling if the strength of the "blockage" is a function of the load. Increased load studies could provide insight into this dependence on load. Since interparticle forces that could contribute to blockage would not be directional, it is not expected that mechanism 1(*b*) could exist except concurrently with mechanism 1(*a*).

Case 1(c)—Selective orientation of anisometric particles will occur during shear if the forces required to randomize the orientation are larger than the shear forces between the oriented particles, *i.e.*, if shear occurs before the particle is turned from its preferred orientation. Consequently, the shear force would be expected to decrease as the orientation occurs. Since bidirectional pulling would disrupt the orientation process, a larger friction coefficient would be observed for the bi- than for monodirectional pulling. M should be positive for mono- and zero for bidirectional pulling. The increased load studies would indicate the dependence of the extent of orientation on the applied load.

Possibly, particle orientation could increase the conformation between particles and a much stronger powder bed would result from the alignment. However, the shearing process must disrupt the bonds to form a region of shear. Probably a thin shear plane would develop and the bed would behave as two compacts sliding over one another.

Case 2—Unfortunately, the plastic deformation contribution to the shear forces cannot be distinguished from the simple consolidation case of 1(*a*) by the procedures outlined here. The same characteristics would be observed as in 1(*a*) unless plastic deformation is not increased by shearing the bed. In this case, $M = 0$ for both mono- and bidirectional pulling. If measurement of bed volume change were made as the powder bed yields to initiate shear, Case 1(*a*) would be accompanied by a dilation of the bed. Dilation need not accompany shear in Case 2. However, the present apparatus does not provide for measurements of bed dilation, and the required sensitivity to make such a measurement useful has not been estimated.

---

[5] $\tau_1 c$ refers to the first pull value of the shear force with applied load equal to c.

[6] Recall that both increased load and reduced load data are obtained after a plateau condition has been established. However, the M, M', μ_1, μ', and $\tau_1 1000$ values depend at least in part on nonplateau conditions of the bed.

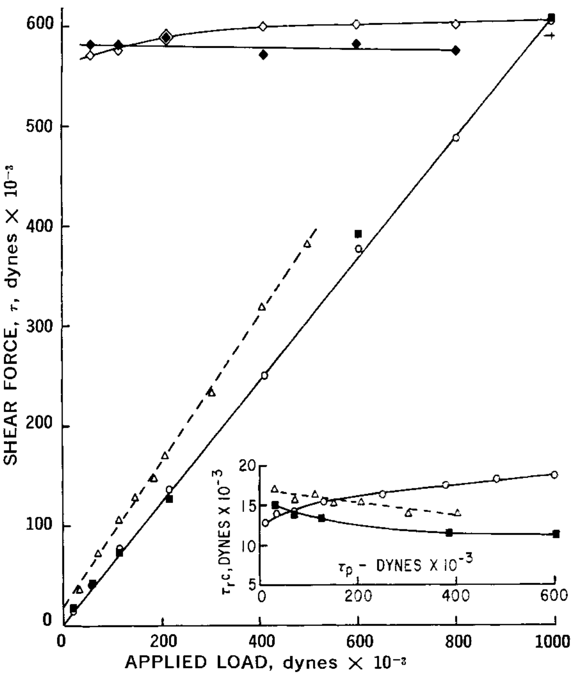

Figure 4—*Spray-dried lactose. Key: main graph:* ⊙, *monodirectional* τ_p, *SPS cell;* ◇, *monodirectional* $x\tau_l 1000$, *SPS cell;* ■, *bidirectional* τ_p, *SPS cell;* ◆, *bidirectional* $x\tau_l 1000$, *SPS cell;* △, *monodirectional* τ_p, *TPT cell, vibrated 1 sec. between each pull;* †, $\tau_l 1000$, *SPS cell; insert graph:* ○, *monodirectional* $x\tau_r 20$, *SPS cell;* ■, *bidirectional* $x\tau_r 20$, *SPS cell;* △, *monodirectional* $x\tau_r 13$, *TPT cell, vibrated 1 sec. between each pull.*

In a previous communication (1), the very large increases in friction coefficient produced by standing with the load applied and also by vibrational forces were illustrated. Relaxation of forces and plastic deformation over long periods is a common phenomenon and, therefore, it is reasonable to assign these shear strength changes to deformation growth of areas of true contact. However it is not simple to prove that no structural changes resulting from particle rearrangement within the bed have occurred.

Chlorphenesin carbamate was shown to double its resistance to shear upon standing with load applied for approximately 24 hr. (1). To produce a comparable effect by a reduced load-type of measurement, one must reach a plateau condition at four times the load if an index of retention of 0.5 applies or at 40 times the load if the index of retention is 0.05.[7] Therefore, if one is to argue that consolidation alone accounts for the increase of shear strength, it appears to be equivalent to the degree of consolidation that would be produced by reaching plateau conditions with 40 times the applied load. Therefore, the authors prefer to assign the changes primarily to deformation. Theoretical aspects relating to plastic deformation have been considered by Krupp (3).

Shear processes with real powders most certainly will involve several of the above processes simultaneously. Unequivocal interpretation may not be possible. However, considerable insight may be obtained. Experimental data obtained with real powders are reported in the following paragraphs.

EXPERIMENTAL RESULTS

A large number of powders of pharmaceutical interest have been investigated in this laboratory. Five examples have been chosen for inclusion here in order to illustrate the variety of results obtained. Some of the measurements described in the preceding paragraphs are illustrated with only one example. However, a minimum set of six plots are given for each—*viz.*, τ_p, $x\tau_l 1000$, and index of retention for both mono- and bidirectional pulling. Some special studies of the effects of milling are included, also. Because these studies have extended over a protracted period of time and various lots of the same chemical compound were used, some apparently disparate values are reported. However, these are believed to be true differences in sample properties. Also, some small variation may be encountered when TPT values are compared with SPS values. It was reported previously that both the experimental procedure and history of the specific sample affect the results of shear cell measurement (1).

Spray Dried Lactose—The values observed with the NF material using the SPS cell and the usual procedures are given in Fig. 4. (For the moment ignore the TPT values for a sample vibrated for one second between each pull that are included in this graph.) The very low values of the index of retention are believed to indicate that the particle-particle interactions are predominantly elastic, *i.e.*, unstressed large areas of true contact do not develop from plastic deformation. This is consistent with the free-flowing properties of this powder. The negative values of the index of retention for the bidirectional case were unexpected. The negative slope could result from either a decreased degree of bed consolidation at plateau conditions with larger loads or from increased disruption of the particle-particle bonds upon removal of the larger loads. The small negative slope of the $x\tau_l 1000_{2d}$ line would be more consistent with the former explanation. The $x\tau_l 1000$, the $\tau_p 1000$, and $\tau_l 1000$ values all fall within a very limited range, so the degree of consolidation doesn't seem to be grossly dependent on the applied load. However, both the $x\tau_l 1000_{1d}$ line and the monodirectional index of retention have small positive slopes. This suggests some small changes with load.

The mono- and bidirectional friction coefficients essentially are equal. Therefore, it is assumed that no structure develops that resists the continuation of motion in the same direction. Two opposing mechanisms producing equal but opposite effects could be occurring but no suggestion of orientation of these irregular-shaped particles is detected and the small consolidation effects make this alternative seem improbable.

From the above observations it is concluded that spray-dried lactose particles undergo predominantly elastic interactions and only minor packing changes occur upon shearing the bed.

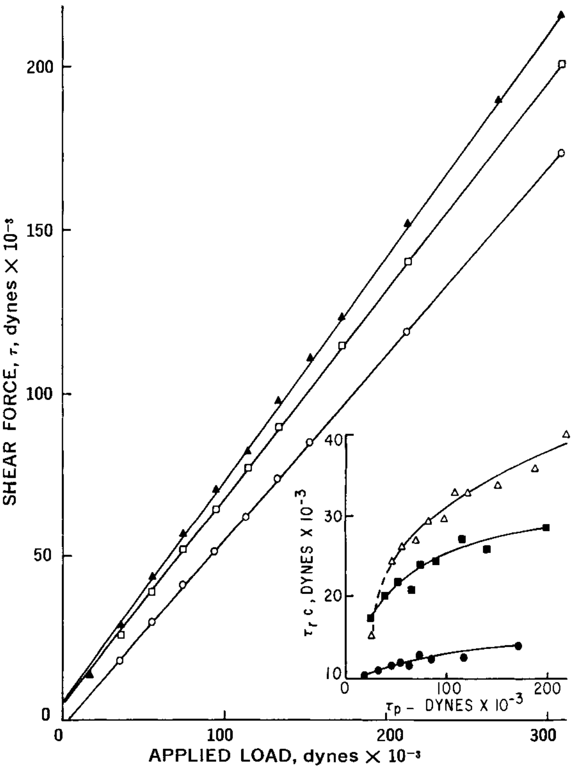

Figure 5—*Spray-dried lactose; monodirectional with TPT cell. Comparison of screened, milled, and unmilled samples. Key: main graph:* ⊙, *unmilled,* τ_p*; diam. <325 mesh;* □, *milled,* τ_p*; diam. >325 <230 mesh;* ▲, *milled,* τ_p*; diam. <325 mesh; insert graph:* ●, *unmilled,* $x\tau_r 10$*; diam. <325 mesh;* ■, *milled,* $x\tau_r 10$*; diam. >325<230 mesh;* △, *milled,* $x\tau_r 10$*; diam. <325 mesh.*

---

[7] The experimental value reported herein is approximately 0.05.

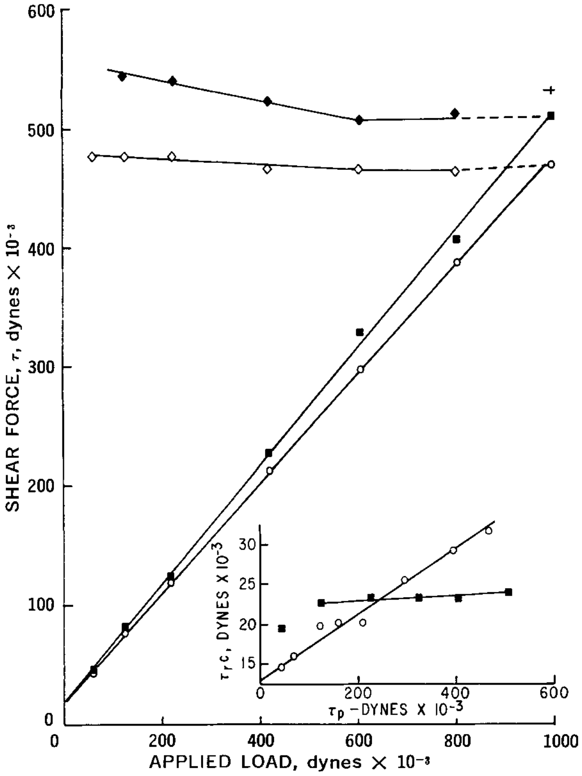

Figure 6—*Chlorphenesin carbamate; SPS cell. Key: main graph:* ○, *monodirectional* τ_p; ◇, *monodirectional* $x\tau_l1000$; ■, *bidirectional* τ_p; ◆, *bidirectional* $x\tau_l1000$; †, τ_l1000; *insert graph:* ○, *monodirectional* $x\tau_r20$; ■, *bidirectional* $x\tau_r20$.

In a special study the powder bed was vibrated by means of an electric vibrator[8] attached to the laboratory jack supporting the TPT cell. The vibration was applied for 1 sec. between each monodirectional pull. The friction coefficient increased to 0.72. The IP pattern changed to Type I-IP with $M = -0.2$; and the index of retention became slightly negative. Possibly, vibration-produced consolidation was inhibited by the larger applied loads. Unfortunately this work was done before the importance of increased load studies was recognized. Therefore, the $x\tau_l1000$ plot was not determined. The reduced load studies do not indicate that plastic deformation is a significant factor. Therefore, the larger friction coefficient of the vibrated material is believed to reflect a higher degree of consolidation.

In still another study with spray-dried lactose, the effects of milling were observed. The sample was milled in a small laboratory ball mill[9] for 6 hr. Selective screenings provided the particle size range desired. Figure 5 compares the results of the TPT cell studies of milled and unmilled material. Because it often has been demonstrated that small particle size contributes to the cohesive properties of powders (4), the authors were particularly interested in the comparison of a milled material screened to pass through a 230 mesh and retained on 325 mesh (⟨230⟩325) screen with unmilled material that passed through a 325 mesh screen. For the milled sample, I-IP characteristics were observed but the smaller particle size unmilled sample retained the P-IP shape. The friction coefficient was changed slightly by milling and the index of retention increased markedly.

The fraction from the mill that was retained on the 120 mesh screen (not shown in Fig. 4) appeared to have passed through the mill unchanged, as judged by the absence of changed in IP, μ, and index of retention values. However, the smallest size fraction of the milled material had an increased friction coefficient and an index of retention several times larger than the unmilled material.

These results do not imply that particle size is unimportant. However, they indicate that other properties also are important. Milling has altered the powder in some manner so that it is less flowable. Probably a change of particle shape, texture, or of surface mechanical properties could produce these results.

Chlorphenesin Carbamate—The SPS cell experimental results with crystalline chlorphenesin carbamate in the form used in pharmaceutical products is shown in Fig. 6. Some TPT cell measurements have been made, also, and the results are shown in Fig. 7. The latter will be discussed first. However, comparison of the monodirectional values indicates only small differences between Figs. 6 and 7, even though different lots of material and different cell types were used for the measurements.

When subjected to shearing action, chlorphenesin carbamate is observed to act as a plastic solid. For example, it is difficult to prevent it from bonding to the die and punch of a tableting machine. Also, difficulty was experienced in milling it at room temperatures. However, compression without shear does not produce strong interparticle bonding. This last observation suggests elastic interactions between particles.

A small sample was ground in a mortar and pestle by mixing it with dry ice. The ground sample had a BET surface area of 4.35 m.²/g., which is larger than the 3.49 m²/g. measured before grinding. It was not fractionated and the size distribution of particles was not determined.

With ground material a true plateau condition was not obtained. The shear force value of the third pull after the peak was selected arbitrarily as representative of the plateau. Probably the error was small because the decrease of shear force became very small after the third pull. Also, this approximates the same number of pulls used to obtain the plateau condition with unground material. Only monodirectional values were determined.

The ground sample P-IP values were $M = 0.2$ and $M' = 0.4$. For unground material they were $M = 0.15$ and $M' = 0.20$. The friction coefficient for the ground sample was determined to be 0.72, considerably larger than the 0.46 value of the unground sample. The index of retention for the ground sample ranged between 0.3 for reduction from small loads to about 0.1 for reduction from larger loads. Both the SPS and TPT data show index of retention values for the unground material to be less than 0.05; in fact, for the TPT data a zero slope was obtained. Qualitatively, the unground powder was a free-flowing powder and the ground material was not.

It would require additional work to establish the cause of the change in properties. However, grinding has either introduced enough fine particle size material to produce this effect or it has altered the surface properties of the solid so that the particle-

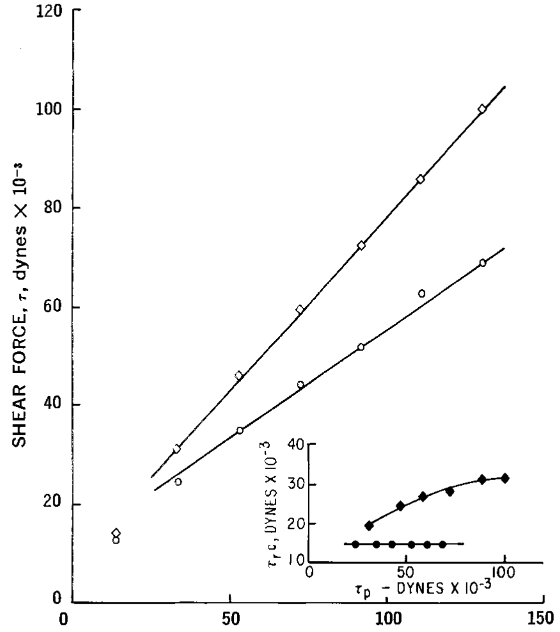

Figure 7—*Chlorphenesin carbamate; monodirectional with TPT cell; change by grinding. Key: main graph:* ○, *unground,* τ_p; ◇, *ground,* τ_p; *insert graph:* ●, *unground,* $x\tau_r14$; ◆, *ground,* $x\tau_r14$.

---

[8] Syntron Electric Vibrator, Type V-4, Style 1518, Syntron Co., Homer City, Pa.

[9] Utility jar mill, United States Stoneware Co., Akron, Ohio.

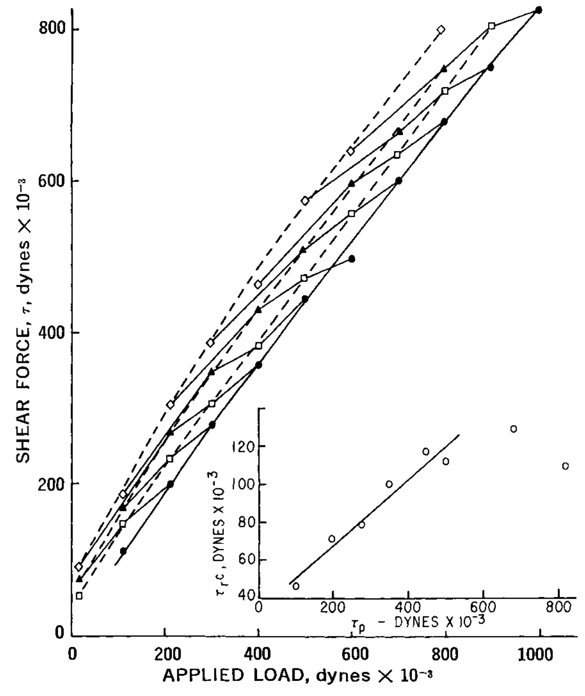

Figure 8—*β-Sitosterol, monodirectional with TPT cell; detailed reduced load shear force data. In the main graph the solid lines to the right of the* τ_p *plot tie together points obtained by reducing the load from a given* τ_p *value. Each dashed line ties together the* $x\tau_{cr}$ *values for a single value of c. Key:* ●, τ_p; □, $x\tau_{100r}$; ▲, $x\tau_{200r}$; ◊, $x\tau_{400r}$; ○, $\tau_p x$ vs. $x\tau_r$*13.*

particle interactions and/or consolidation have increased. The latter explanation would be consistent with both these results and those for lactose. Of course, both factors could be contributing to the large change in properties of this material.

In Fig. 6 are shown both mono- and bidirectional SPS friction plots, increased load shear forces, and reduced load results. Note that the bidirectional friction coefficient is larger and the index of retention values smaller than the respective monodirectional values. The increased load, $x\tau_I1000$, shear lines indicate that the shear-induced conditioning is dependent on the load at smaller loads and becomes independent of the load at larger loads. For the smallest loads the bidirectional $x\tau_I1000$ values are between the $\tau'1000$ and τ_I1000 values.

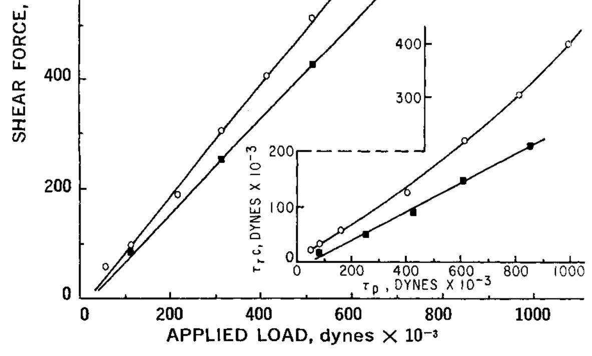

Figure 9—*β-Sitosterol, SPS cell. Key: main graph:* ○, *monodirectional* τ_p; ◊, *monodirectional* $x\tau_I$*1000;* ■, *bidirectional* τ_p; ◆, *bidirectional* $x\tau_I$*1000;* †, τ_1*1000; insert graph:* ○, *monodirectional* $x\tau_r$*20;* ■, *bidirectional* $x\tau_r$*20.*

The IP values are $M_{1d} = 0.14$, $M'_{1d} = 0.10$, $M_{2d} = 0.057$, and $M'_{2d} = 0.10$. These M values confirm the conclusions based on the increased load values, *i.e.*, they indicate that conditioning occurs with both mono- and bidirectional pulling and that the conditioning is different for the two cases. The very low index of retention values indicate that neither consolidation nor plastic deformation is contributing grossly to the shear-induced conditioning.

The above observations suggest that the particle-particle interactions are predominantly elastic and that the orientation of particles is largely responsible for the changes in shear force and for the differences between mono- and bidirectional values. This effect must overcome any tendency to develop structure that resists the continuation of motion in the same direction. Otherwise, $\mu_{2d} < \mu_{1d}$. Microscopically, platelet crystals are observed.

The cause of differences between the TPT and SPS value for both M'_{1d} and the index of retention are not obvious. With the TPT arrangement the peak shear force usually was reached on the second pull. However, with the SPS arrangement, four or more pulls were required to reach a peak value. Also, the area of the top plate is smaller in the TPT cell. Possibly some of these factors have a small influence on the results.

β-Sitosterol—Powdered β-sitosterol was prepared by mixing coarse material (NF grade) with crushed dry ice and passing the mixture through a hammer mill.[10] The TPT data in Fig. 8 were obtained with a different lot of material than the SPS data in Fig. 9. Because this powder has a large index of retention, it is an excellent material for illustrating more completely the retention of shear strength after reduction of applied load. In Fig. 8 the $c\tau_rx$ values are connected by solid lines appearing as branches to the left of the τ_p plot. The same points are used for the $x\tau_cr$ lines shown connected with dashed lines.

The extra retained shear strength acts only on the first pull. On the second pull after reducing the load the shear strength is essentially equal to the plateau value at the reduced load.

The data in Fig. 9 provide additional insight into the properties of these powder beds. $M_{1d} = -0.44$ and $M_{2d} = -0.32$; also the index of retention values are >0.2 over the entire range of loads studied. The increased load lines indicate that the extent of shear-induced conditioning increases nearly linearly with load for the monodirectional case but undergoes decreased rate of change in conditioning at larger loads with bidirectional pulling.

Previously the authors (1) reported on the increase of friction coefficient of spray-dried lactose and chlorphenesin carbamate produced by moderate levels of vibration. Use of the same procedure with β-sitosterol produced no detectable increase of friction coefficient. Possibly the magnitude of forces produced by the vibration was inadequate to overcome the strong bond between particles.

The above observations suggest that either the interactions between the particles are predominantly plastic or that consolidation proportional to applied load occurs. Differences between mono- and bidirectional results indicates that orientation and/or structure develops that resists resumption of motion in the same direction. However, the latter effects are the dominant ones in determining the flow properties of this powder.

The large friction coefficient for this material suggests strong interactions between the particles. However, it may result either from weaker interactions at large areas of true contact or stronger interactions at smaller areas of true contact. Consolidation must be accompanied by an increase in contact area to produce such a large friction coefficient. The authors conclude that the evidence for plastic deformation producing large areas of true contact is dominant but not unequivocal for this material.

Calcium Carbonate—Of interest, also, are the results of measurements made with a precipitated calcium carbonate that was judged, qualitatively, to have poor flowability. This material was of special interest because it is the only material discussed here that did not bond under pressure to form a compact of sufficient strength to be removed intact from the die. Therefore, it was assumed that the calcium carbonate was an elastic solid undergoing only limited

---

[10] Mikro Pulverizer, Type CF, Pulverizing Machinery Div., Metals Disintegrating Co., Inc., Summit, N. J.; 0.025-cm. (0.010-in.) herringbone screen.

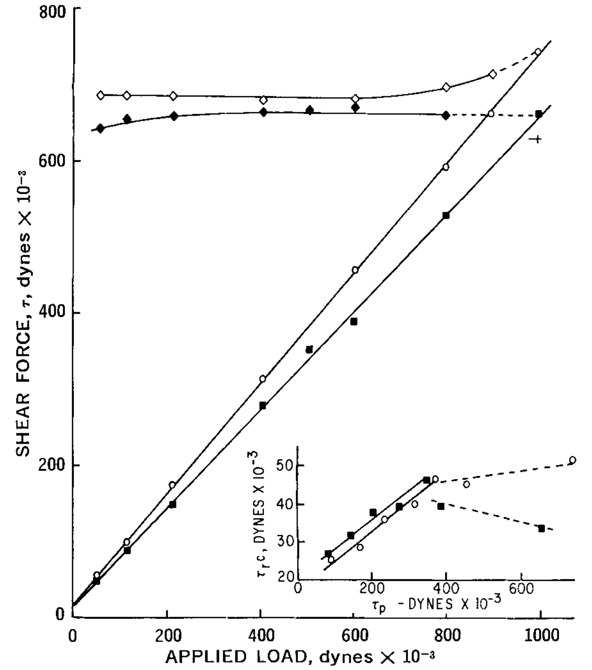

Figure 10—*Calcium carbonate; SPS cell. Key: main graph:* ○, *monodirectional* τ_p; ◇, *monodirectional* $x\tau_I 1000$; ■, *bidirectional* τ_p; ◇, *bidirectional* $x\tau_I 1000$; †, $\tau_I 1000$; *insert graph:* ○, *monodirectional* $x\tau_r 20$; ■, *bidirectional* $x\tau_r 20$.

plastic deformation under compression forces larger than normal for the shear cell.

The results are shown in Fig. 10. $M = -0.1$ for monodirectional, -0.03 for bidirectional pulling. The latter procedure produces a P-IP pattern with $M' = 0.05$. Hence less shear-induced conditioning occurs with bidirectional pulling. The unusual index of retention plots show large, abrupt changes from a value large enough to indicate poor flowability to a value associated with good flowability. The nearly flat increased load line indicates that this slope change is not due to differences in the shear-induced conditioning in the load range studied. Also, the similarity of the index of retention values for mono- and bidirectional pulling show that the effect is nearly direction independent. The difference in the monodirectional, 0.73, and the bidirectional, 0.59, friction coefficients indicate some form of direction-dependent resistance to shear. Also, the M values show that the shear-induced conditioning is more extensive for the monodirectional pulling. Particle orientation could account for a change in both μ and the index of retention. However, the index of retention remains approximately the same. Therefore, it is hypothesized that the calcium carbonate shear-induced conditioning is not primarily particle orientation but the development of structure that resists continuation of motion in the same direction. It seems that consolidation and/or plastic deformation of surface contact regions occurs at small loads with elastic deformation becoming dominant at larger loads.[11] Probably plastic deformation contributes significantly to the conditioning at small loads since the mono- and bidirectional index of retention values are equal and large in this region but the increased load lines are nearly flat.

Magnesium Stearate—Portions of Fig. 11 correspond to Fig. 13 of *Reference* 1 and show anomalous friction plots obtained with magnesium stearate. Here, both the 200 $\tau_r x$ and the 1000 $\tau_r x$ lines obtained with the TPT cell have been added. As indicated previously, the regions of decreased shear forces are believed to be the result of shear-induced conditioning of the tablet in TPT studies and of caking onto the sandpaper followed by similar conditioning for the SPS studies. With the SPS cell anomalous results occurred only with load in excess of 1000 g. In Fig. 11, note that when the load is reduced from a value above the inflection point, the $c\tau_r x$ line is below the friction plot. However, when reduced from a load value less than the anomalous region, a typical reduced load plot is obtained. This confirms that irreversible changes are occurring with shear and larger applied loads. Perhaps this anomalous region results from the same property of magnesium stearate that accounts for its lubricant properties in tablet dies.

Since the onset of the anomalous results with the SPS cell occurs at loads in excess of 1000 g., results comparable to those reported

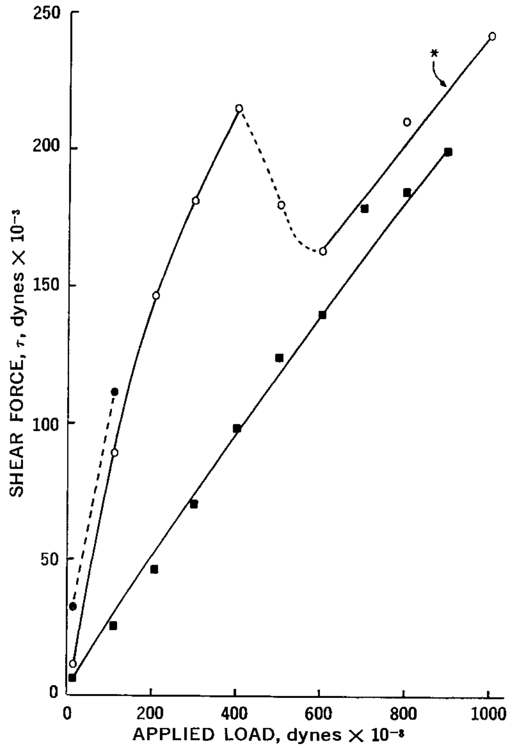

Figure 11—*Magnesium stearate, monodirectional with TPT cell. Key:* ○, τ_p (**slope based on additional data not shown in this graph*); ●, *200* $\tau_r x$; ■, *1000* $\tau_r x$.

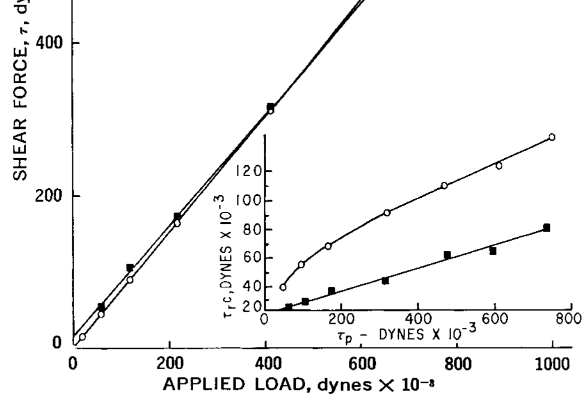

Figure 12—*Magnesium stearate, SPS cell. Key: main graph:* ○, *monodirectional,* τ_p; ◇, *monodirectional,* $x\tau_I 1000$; ■, *bidirectional,* τ_p; ◆, *bidirectional,* $x\tau_I 1000$; †, $\tau_I 1000$; *insert graph:* ○, *monodirectional,* $x\tau_r 20$; ■, *bidirectional* $x\tau_r 20$.

---

[11] The relationship of true contact area to volume of solid stressed elastically is an important consideration in determining whether elastic rebound or removal of load will break the "bond" between particles. An analogy with the critical load in burnishing and polishing might be useful (5).

for the other powders could be obtained. These are shown in Fig. 12. The friction coefficients for mono- and bidirectional pulling are 0.76 and 0.74, respectively. The patterns are I-IP for monodirectional pulling, $M = -0.13$ and P-IP for bidirectional pulling, $M = -0.12$ and $M' = 0.01$. Therefore, the consolidation of the powder with bidirectional pulling must occur either at a different rate or by a different route than with monodirectional pulling. Even though the friction coefficients (μ_p) are essentially equal, the monodirectional and bidirectional plateau conditions seem to be different conditions of the bed since the reduced load lines, $x\tau_r c$ are quite different. The increased load line shows the degree of consolidation to be dependent on the applied load. The index of retention obtained by reduction from lower loads indicates that the magnesium stearate will not have good flow properties. However, the retention effect is larger for inducing shear in the same direction than for the reversed direction of pull.

CONCLUSIONS

Although the results of this extended series of studies provides only limited evidence of the mechanics of powder shear, the study is believed by the authors to provide extensive, new insight into differences among powders and to provide new parameters other than friction coefficient to express these differences. These parameters are believed to indicate whether or not a powder will flow readily and also to indicate the changes that may occur as the forces normal to the shear plane are varied. Additional work must be done to show whether or not quantitative correlations with flowability under specific conditions can be established.

NOMENCLATURE

IP = Initial to plateau shear forces used in describing patterns obtained in the series of pulls from first pull to plateau conditions.

I-IP; D-IP; and P-IP = Increasing, decreasing, and peaking (showing a maximum), respectively, shear forces in the IP patterns.

μ = Friction coefficient based on Eq. 1.

μ_1, μ_p, and μ' = First pull, plateau, and peak values, respectively, of friction coefficient.

M = (Capital letter μ) defined by Eq. 2 and expresses quantitatively the change in shear force from the initial to plateau pull.

M_{1d} and M_{2d} = M value calculated using data obtained with monodirectional and bidirectional (alternating direction) pulling, respectively.

M', ${}_{1d}'M$ and ${}_{2d}'M$ = Analogous to corresponding M values (see Eq. 3) but compare peak values of shear force with plateau condition values.

τ = Shear force.

τ_1, τ_p, and τ' = Shear force for first pull, plateau, and peak value (for P-IP case), respectively.

τ_{p1d} and τ_{p2d} = Plateau condition shear force for monodirectional and bidirectional pulling, respectively.

$\tau_p c$ and $\tau' c$ = Corresponding shear forces for a specific value, c, of the applied load. c may be specified in grams, *e.g.*, $\tau_p 1000$.

τ_r = A reduced load shear force.

τ_{r1d} and τ_{r2d} = A τ_r value obtained with monodirectional or bidirectional pulling, respectively.

$c\tau_r x$ = A series of reduced load shear force values, each obtained from the same plateau condition, $\tau_p c$, by reducing the load by different amounts, x. The c to x order implies that the plateau condition load was held constant, c, for all measurements but the load after reduction, x, was varied. The symbol could be considered a contraction of symbols for a two-step process, *i.e.*, $\tau_p c - \tau_r x$ contracted to $c\tau_r x$.

$x\tau_r c$ = A series of reduced load shear force values, each obtained by always reducing the load to the same value, c, from various plateau conditions, $\tau_p x$. The symbol could be considered a contraction of symbols for a two-step process, *i.e.*, $\tau_p x - \tau_r c$ contracted to $x\tau_r c$.

$x\tau_{cr}$ = A series of reduced load shear force values in which each value of the series is obtained by reducing the load by a constant amount (*cr* for constant reduction) after obtaining a plateau condition. Obviously various applied loads must be used to reach plateau conditions for this to become a series of values.

τ_I = An increased load shear force; the shear force observed for the first pull after increasing the load from a value used to obtain a plateau condition.

τ_{I1d} and τ_{I2d} = τ_I values for monodirectional and bidirectional pulling, respectively.

$x\tau_I c$ = A series of increased load shear force values, each obtained by always increasing the load to the same value, c, from various plateau conditions, $\tau_p x$. This symbol could be considered a contraction of symbols for a two-step process, *i.e.*, $\tau_p x - \tau_I c$ contracted to $x\tau_I c$.

REFERENCES

(1) E. N. Hiestand and C. J. Wilcox, *J. Pharm. Sci.*, **57**, 1421 (1968).

(2) J. C. Williams and A. H. Birks, *Powder Technol.*, **1**, 199 (1967).

(3) H. Krupp, *Advan. Colloid Interface Sci.*, **1**, 111(1967).

(4) R. Farley and F. H. H. Valentin, *Powder Technol.*, **1**, 344 (1967/68).

(5) E. Rabinowicz, *Sci. Am.*, **218**, 91(1968).

ACKNOWLEDGMENTS AND ADDRESSES

Received April 11, 1969 from the *Pharmacy Research Unit, The Upjohn Company, Kalamazoo, MI 49001*

Accepted for publication July 31, 1969.

The authors wish to thank Mr. Jon Swanson, Mr. Sydney Jackson, and Mr. John Bane for making many of the measurements reported herein.