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COMPRESSIONAL BEHAVIOUR OF AN AGGLOMERATED CELLULOSE POWDER

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ABSTRACT

The ability of an agglomerated cellulose powder to total and plastic deformation was evaluated and compared with those of Avicel PH 101, Emcocel and an experimental depolymerized cellulose powder. The elastic recovery of compressed cellulose tablets was also measured. The effects of deformation of the material during the tableting process and recovery of tablet after maximum compression on the mechanical strength of tablets were also discussed.

The apparent net work done into tablets during compression as well as the yield pressures to total and plastic deformation, determined from the Heckel treatment, showed no great differences between the agglomerated cellulose powder and the other cellulose powders. Thus all the cellulose materials studied had rather similar ability to total, i.e. elastic and

plastic, deformation and to permanent, i.e. pure plastic, deformation. The obvious fragmentation of the agglomerated cellulose powder already at low compressional pressure, however, seemed to be advantageous for the formation of strong compacts.

Both rapid and total elastic recovery of compressed cellulose tablets showed clear differences between the cellulose materials and these differences correlated with the previously measured strength of cellulose tablets. The agglomerated cellulose powder had the smallest tendency to both kind of elastic recoveries of tablets. Obviously, due to the large interparticle contact areas, the ability of this material to establish more bonds between adjacent particles during compression was greater than those of other celluloses. The elastic recovery was greatest for depolymerized cellulose tablets indicating the poorest binding ability of the particles of this material.

INTRODUCTION

The ability of a material to permanent deformation during compression is a prerequisite for the formation of a compact. Deformation of particles allows them to come into a contact close enough for bond formation. Several different methods have been used to evaluate the deformation properties of tablet excipients. One of the most often used method is the evaluation of porosity changes as a function of compressional pressure, usually using the so called Heckel plots (1,2,3). Other methods employed include stress-relaxation studies (4), work measurements from force-displacement data (5), surface area changes during compression (6) and scanning electron microscopy (7). All these methods have their own advantages and

disadvantages. Thus they are not exclusive, but complementary to each other. Besides the deformation of a material during compression also the behaviour of the compressed tablet during decompression and ejection and also after ejection of the tablet from the die affects the ability of the material to form strong compacts. Aulton et al. (8) pointed out that materials which deform plastically with little elastic recovery should produce better quality tablets than more resilient materials. Duberg and Nyström (1) suggested that the possibility of certain points on particle surfaces to establish the interparticle attraction would be more important for bonding of the material than the deformation of powder during compression.

The ability of microcrystalline cellulose to plastic deformation is well reported (4,9,10). The random distribution of microcrystalline particles in the cellulose aggregates allows for plastic deformation because of the presence of slip planes and dislocations on microscale, and deformation of the aggregate on macroscale, which all make microcrystalline cellulose powder extremely compressible (9).

One of the few problems associated with microcrystalline cellulose is its poor flowability. The commercial, partially granulated, form of microcrystalline cellulose, Avicel PH 102, with larger particle size than mostly used commercial trade Avicel PH 101, provides somewhat better flowability without a significant loss in compressibility (9). However, the flowability problems are not totally overcome by the use of Avicel PH 102 instead of PH 101. Staniforth et al. (11) studied the effect of moisture addition on the properties of two microcrystalline celluloses, Avicel PH 101 and Emcocel. They observed a dramatic decrease in compactability of both microcrystalline cellulose samples when they were wet granulated. According to them improved flowability and increased compactability were exclusive.

Pesonen et al. (12) have reported the excellent tableting properties of a novel agglomerated cellulose powder, these including both better flowing as well as binding properties than those of microcrystalline celluloses, Avicel PH 101 or Emcocel. More recently the effects of crystal, particle and powder properties of this agglomerated cellulose powder and three other cellulose powders on the strength of compressed tablets were evaluated (13). Clear differences in the strength of tablets compressed at 65 MPa were observed (13). The breaking strength of depolymerized cellulose tablets was only one third and the breaking strength of Avicel PH 101 and Emcocel tablets two thirds of the breaking strength of agglomerated cellulose tablets. According to that study the ability of these four celluloses to produce strong compacts correlated especially with the amount of specific surface area of the cellulose powders. There existed, however, also a clear difference in crystallinity between the agglomerated cellulose powder and the other three celluloses, which could lead to a difference in their deformability and thus also in the ability to form strong compacts.

The aim of this study was to evaluate the deformation behaviour of the agglomerated cellulose powder during tableting process and the elastic recovery of corresponding tablets. These properties were compared with those of three other cellulose powders and the possible differences between celluloses were discussed with the concept of their effect on the mechanical strength of compacts.

MATERIALS AND METHODS

Materials

The cellulose materials studied were the agglomerated cellulose powder (ACP), a depolymerized cellulose powder (DCP) and two

commercially available microcrystalline cellulose powders, Emcocel and Avicel PH 101. The first three materials were supplied by Finnish Sugar Ltd (Kantvik, Finland) and the fourth was manufactured by FMC Corp. (Philadelphia, USA). The crystal, particle and powder properties of these materials have been discussed previously (13).

Methods

Tablets were compressed using an instrumented Korsch EK-O single punch machine with flat faced 13 mm punches. Compressional forces were recorded using strain gauges and upper and lower punch movements using inductive displacement transducers. The machine speed used was 35 tablets per minute. Tablets were compressed from separately pre-weighed 300 mg amounts of plain cellulose powders. The apparent particle densities for the celluloses studied were nearly the same, 1.50 - 1.52 g/ml (13). Thus also the true volume of powder poured into the die was nearly the same in every case.

The net work done into tablets during compression was calculated by subtracting the expansion work and the friction work according to Järvinen and Juslin (14) from the upper punch work.

The deformation properties of cellulose powders were studied using the Heckel equation (15,16)

$$\ln\left(\frac{1}{1 - D}\right) = kP + A \qquad (1)$$

where D is the apparent density of tablet at the pressure P

divided by the apparent particle density of the material to be compressed. P is the applied pressure and k is the slope of the straight line portion of the curve. The reciprocal of k is referred as the yield pressure k_p. Tablet dimensions, for calculation of Heckel values were measured 24 hours after compression. Tablet dimensions were measured also one week after compression but no further expansion of tablets was seen. Thus k_p describes purely the tendency of the materials to plastic deformation. The dimensions of the powder column were also determined at maximum compression. The obtained k_d value includes besides plastic, also elastic component of deformation.

The value of D_A, corresponding the intercept A of Heckel plot, describes the densification of powder due to both die filling and rearrangement of particles. The value of D_0 is obtained by dividing the bulk density of the powder with the apparent particle density and describes the densification due to filling of the die. The difference between D_A and D_0, called D_B, describes the densification of powder due to particle rearrangement.

Scanning electron micrographs taken from both broken and upper surface of tablets were used to evaluate visually the deformation of the materials. Micrographs were taken with Jeol JSM-35 apparatus.

The elastic recovery of tablets was calculated using the equation

$$\text{Elastic recovery} = \frac{h_t - h_{max}}{h_{max}} \times 100\ \% \qquad (2)$$

where h_{max} is the height of powder column at maximum

compression and h_t is tablet height either immediately, 24 hours or one week after ejection of the tablet from the die.

To get an impression of the rapid elastic recovery of the cellulose materials after the removal of load, the apparatus originally constructed for determination of elastical properties of cartillage (17), was used. The load of 20 g was transmitted to tablet upper surface through a cylindrical plane-ended steel intender with a diameter of 400 um. The load was removed after 15 seconds loading and elastic recovery of tablet during next two seconds, under the 1 g tare load of the intender, was measured. The measurement was carried out after the elastic recovery of tablet after compression was totally over. The values presented are the means of five determination points across the tablet surface. The value of each determination point is a mean of three tablets.

RESULTS AND DISCUSSION

Apparent Net Work

Ragnarsson and Sjögren (3) have pointed out that net work done into tablets is greatly affected by particle interactions during compression and is a poorer measure for the plastic properties of the material than the yield pressure of Heckel plots. The differences in apparent net work between the cellulose materials were only small (Fig. 1) and no clear correlation with the previously measured tablet strength was observed. The slightly greater net work of ACP tablets could possibly be due to the amount of work consumed for the fragmentation of large agglomerates into smaller 'primary' particles.

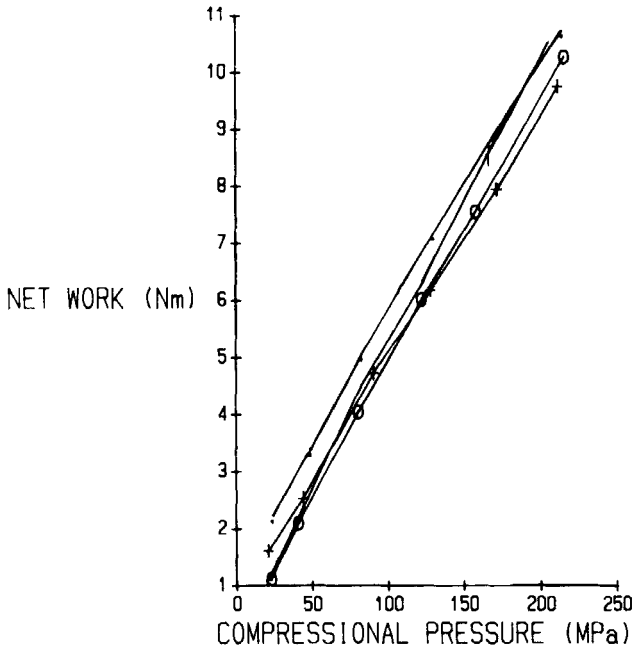

Figure 1.

Net work done into cellulose powders during compression. Avicel PH 101 (—), Emcocel (⊖), ACP (⊤) and DCP (+). The bars indicate standard error of the mean (if excluded they are falling within symbols).

Deformation Properties Based on Heckel Equation

The shapes of Heckel plots for ejected tablets were quite similar for all the celluloses (Fig. 2). The reciprocal of the slope of the straight line portion i.e. the yield pressure k_p, calculated using the values between 20 and 220 MPa was about the same for ACP and Avicel, being slightly smaller than the values for Emcocel and DCP (Table 1). Also the Heckel plots obtained using the tablet in-die-method were rather similar for the celluloses (Fig. 3). The yield pressure k_d, calculated using the values between 20 and 145 MPa, was about the same for

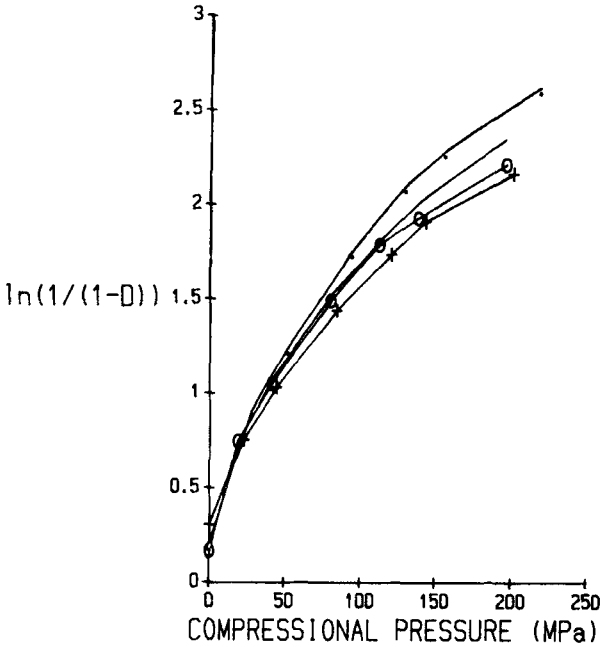

Figure 2.
Heckel plots for ejected tablets. Key as in Fig. 1.

Table 1. The yield pressures k_p and k_d with the coefficient of correlation of linear regression and D_A-, D_0- and D_B-values for the cellulose powders.

	k_p(MPa) (r)	k_d(MPa) (r)	D_A	D_0	D_B
Avicel PH 101	111.2 (0.986)	57.3 (0.993)	0.505	0.172	0.333
Emcocel	120.2 (0.977)	55.8 (0.989)	0.513	0.153	0.360
ACP	110.2 (0.981)	51.4 (0.973)	0.545	0.184	0.361
DCP	124.9 (0.982)	56.3 (0.985)	0.494	0.263	0.231

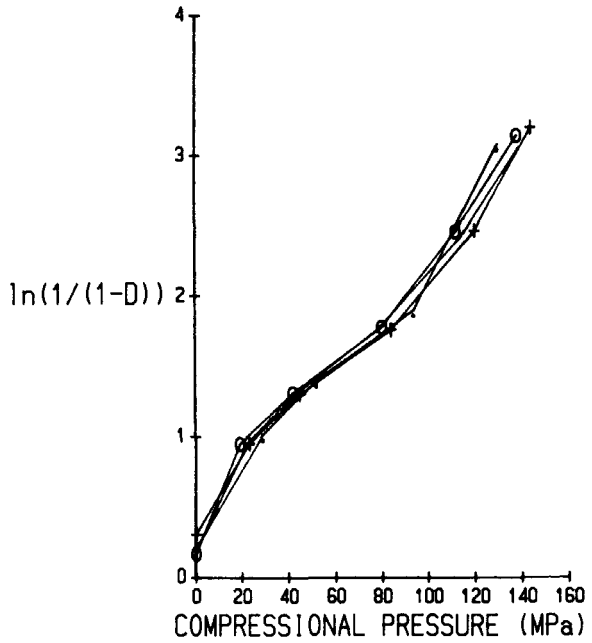

Figure 3.
Heckel plots for tablets-in-die-method. Key as in Fig. 1.

Avicel, Emcocel and DCP and only slightly smaller for ACP (Table 1).

The k_p-value is related to plastic, permanent deformation of the material, which occurs by the means of either plastic flow or fragmentation of particles. The value of k_d reflects the tendency of the material to total deformation including also the elastic component. ACP was thus slightly most prone to both total and permanent deformation and also Avicel was slightly more prone to permanent deformation than Emcocel and DCP. A possible reason for the small difference in deformation behaviour for the advantage of ACP could be the most amorphous structure of this material (13). Thus there could be more lattice defects in the structure of this material accounting for the better deformation properties. The differences were,

however, as in the case of net work done into tablets quite small. Thus the ability of the cellulose powders to undergo deformation was according to yield pressures quite similar and could not be a primary reason for the clear differences observed in the ability of the celluloses to form strong compacts (13).

Duberg and Nyström (1) have presented a theory concerning the volume-reduction of pharmaceutical materials, which often consist of aggregates of 'primary' particles and could be highly porous. These 'secondary' particles could during the initial loading behave mainly as brittle units, with a neglible ability to plastic deformation. After initial fragmentation of aggregates the formed 'primary' particles would, when compressional pressure is increased, deform elastically, plastically or undergo fragmentation. This theory seems to apply well to the behaviour of ACP during compression. This material consists of very porous agglomerates of small cellulose particles resulting to a specific surface area 50 times greater than those of Avicel and Emcocel (13). It is well possible that these agglomerates underwent fragmentation at low compressional pressures thus forming smaller plastically deforming cellulose particles. This suggestion is in agreement with the scanning electron micrographs taken from the broken surface of tablets compressed at the pressure of about 75 MPa (Fig. 4). The original boundaries of large and spherical ACP agglomerates had mostly disappeared indicating a fragmentation of these agglomerates. On the contrary, the particle boundaries could be clearly seen especially in DCP tablets but also in Avicel and Emcocel tablets. Also the scanning electron micrographs taken from the upper surface of tablets compressed at low pressure of 15 MPa (Fig. 5) agreed with the observations from Figure 4 and showed that ACP agglomerates had deformed most effectively and partially lost their identity. From Figure 5 it is obvious that permanent deformation, at least in some extent, occured at clearly lower pressures than

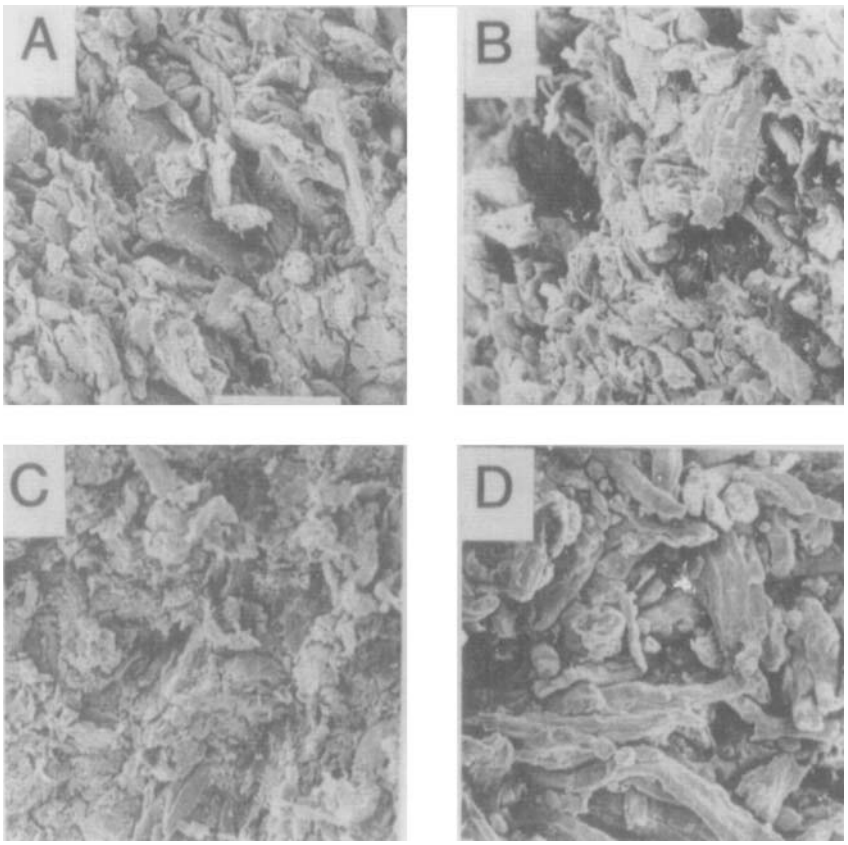

Figure 4.
Scanning electron micrographs taken from the broken surface of tablets compressed at 75 MPa. Avicel PH 101 (A), Emcocel (B), ACP (C) and DCP (D). The bar is 100 um.

indicated by the yield pressure. Paronen and Juslin (18) have pointed out that yield pressure is not an exact limit value but describes the tendency and ability of the material to undergo deformation. The k_p-values for ACP and Avicel were about the same and these materials should thus be equal prone to permanent deformation. k_p is determined from the reciprocal of

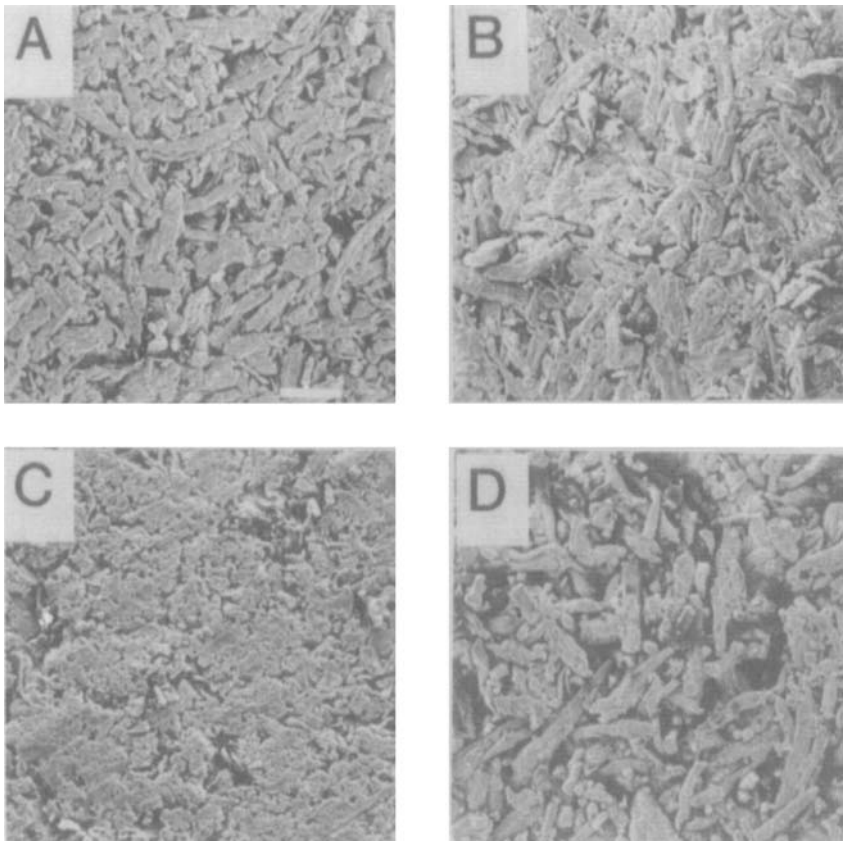

Figure 5.
Scanning electron micrographs taken from the upper surface of tablets compressed at 15 MPa. Key as in Fig. 4. The bar is 100 um.

the straight line portion of Heckel plots and the values before the beginning of the straight line portion are excluded in the calculation. It is possible that the fragmentation of ACP agglomerates at low compressional pressures led to more effective densification before the straight line portion. This on its behalf decreased the slope of the Heckel plot of ACP and thus increased the observed k_p-value. This suggestion was

supported by the values of D_B and D_0 (Table 1). According to D_0-values the rank order for densification of cellulose powders due to die filling was DCP, ACP, Avicel and Emcocel. It could be thus expected that densification due to particle rearrengement after die filling, D_B, would have been greatest for Emcocel and Avicel. However, ACP had the greatest D_B-value. This was obviously due to the partial fragmentation of ACP agglomerates during the initial stages of compression, which resulted to a denser packing of ACP particles.

Elastic Recovery of Tablets

The elastic recovery was clearly smallest for ACP tablets (Fig. 6). The difference between ACP and the other cellulose tablets was most pronounced at small compressional pressures. This could be due to the earlier mentioned fragmentation of ACP agglomerates, which allowed the particles to come into much closer contact than was possible in other cellulose tablets. The deformation properties of cellulose powders were according to yield pressures quite similar (Table 1) and the attraction energy per unit area of true contact is of the same order of magnitude for nearly all organic materials, thus also for cellulose powders (19). Because of both large specific surface area and fragmentation of agglomerates, bringing the formed 'primary' particles into a close contact, a great number of bonds was formed between the particles and elastic recovery of ACP tablets was smaller than those of other cellulose tablets. The observed elastic recovery of tablets correlated well with both spesific surface area of cellulose powders as well as with breaking strength of cellulose tablets (13).

All the cellulose tablets showed minimum elastic recovery at about 80-100 MPa compressional pressure (Fig. 6A and B). The differences between the materials were, however, clear at this point and correlated well with the scanning electron

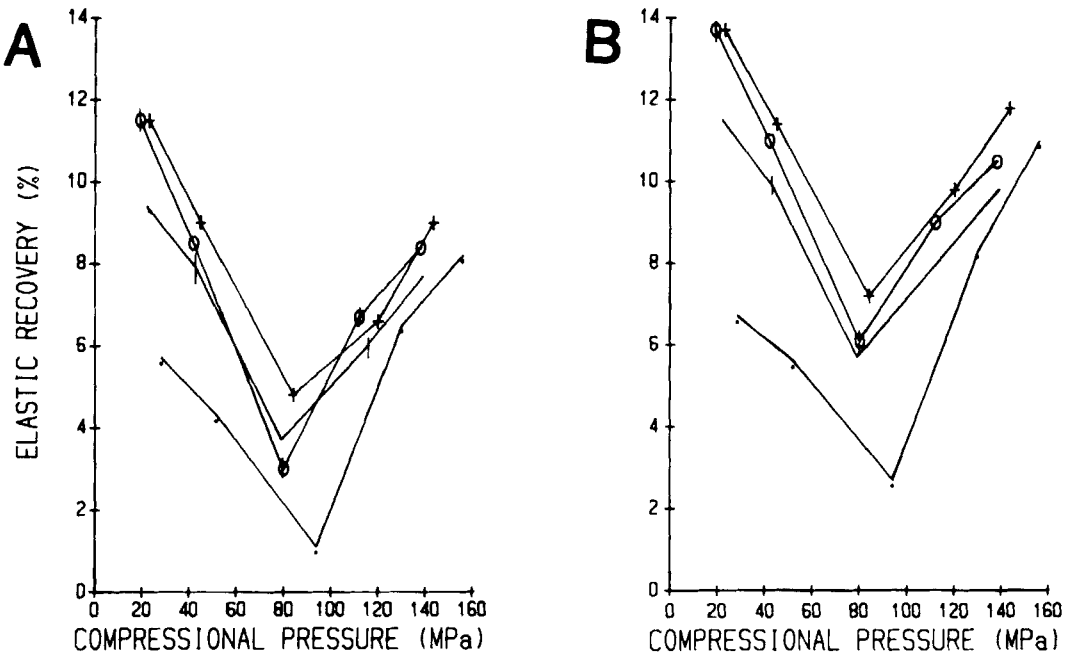

Figure 6.

The slow elastic recovery of tablets immediately (A) and 24 hours (B) after the ejection of the tablet as a function of compressional pressure. Key as in Fig. 1. The bars indicate standard error of the mean (if excluded they are falling within the symbols).

micrographs taken from the broken surface of tablets compressed at 75 MPa (Fig. 4). The broken surface of DCP tablets was clearly loosest showing only moderate interparticle bonding compared to bonding in other cellulose tablets. Patel et al. (20) suggested that extensive and strong interparticle bonding should cause the fracture rather across the microcrystalline cellulose particles than around the boundaries of particles.

This suggestion, however, disagreed with the visual examination of broken surface of both Avicel and Emcocel

tablets (Fig. 4). The fracture seems to have occured mainly around the particles. In ACP tablets, however, the particle boundaries were not so clearly visible and according to the concept of Patel et al. (20) the interparticle bonding was strongest in these tablets.

At higher compressional pressures the elastic recovery started to increase after the minimum value at about 80-100 MPa and also the differences between cellulose materials became smaller. At higher compressional pressures greater amount of energy produced is consumed to purely elastic deformation of the cellulose compact (21). Celik and Travers (22) found that at higher pressures Avicel tablets became more plastic and produced stronger interparticle bonding. However, at high pressures also the elastic recovery of tablets increases. Celik and Travers developed a parameter, which took into account both these effects. They showed that too high compressional pressure is disadvantageous for formation of tablets. Thus to avoid unnecessary and also harmfull elastic work the optimal compressional pressure for plain cellulose powders would according to the results of this study be about 80-100 MPa.

The total elastic recovery of cellulose tablets in Fig. 6B showed a similar kind of trend between cellulose tablets as the elastic recovery immidiately after the ejection of tablets. The elastic recovery of tablets during 24 hours after ejection was smallest for ACP tablets (Fig. 6A and B). However, the elastic recovery of tablets after ejction was rather similar for all the cellulose tablets and ceased in all cases during 24 hours.

Rapid Elastic Recovery of Tablets

The elastic recovery calculated from equation 2 is perhaps the most widely used criterion in quantifying the expansion of

formed tablets. According to Krycer et al. (23) the most logical method for evaluating the elastic recovery is to plot the axial recovery of tablets after ejection, calculated from equation 2, against energy of compaction. Celik and Travers (22), however, pointed out that elastic recovery measured after ejection includes both a rapid expansion of tablet as well as a slower viscoelastic recovery of tablet. They showed, using a hydraulic testing press, that rapid elastic recovery of tablet occurred in 100 ms during decompression. Krycer et al. (23), on the other hand pointed out that the registration of expansion of tablet during decompression in a tablet machine is often incomplete due to the differences in speed between the ascending upper punch and the expanding tablet. This difference should be most pronounced for cohesive materials, such as cellulose powders. In this study the rapid elastic recovery of tablet during next two seconds after removal of load was evaluated in a special apparatus. The test was carried out using compressed tablets and thus resembles in some extent the use of second compression to evaluate the elastic properties of tablets. The situation in the test procedure is, however, not similar as in a tablet machine. Also the recording time of two seconds is longer than that used by Celik and Travers (22). However, the problem of different speeds of ascending punch and expanding tablet could be avoided and the evaluation time of the rapid elastic recovery after load removal is significantly shorter than that obtained when recovery is recorded immediately after ejection. The measurement area on the surface of tablet was small and the measurement must be considered localized. However, five points across the tablet surface were measured in three different tablets. The values in Table 2 are thus means of fifteen measurement points and should be representative for a tablet as a hole. We thus found this method to give a reasonable comparison parameter for the rapid elastic recovery of cellulose tablets.

Table 2. Absolute (um) and relative (%) rapid elastic recovery of compressed cellulose tablets with the standard error of the mean.

	30 MPa compressional pressure	90 MPa compressional pressure
Avicel PH 101	9.0(1.3) um	3.7(0.3) um
	26.4(2.4) %	18.6(1.5) %
Emcocel	12.7(1.2) um	4.0(0.3) um
	29.2(2.4) %	22.8(1.8) %
ACP	4.9(0.5) um	2.4(0.2) um
	19.5(1.3) %	17.5(1.7) %
DCP	17.3(0.8) um	6.4(0.9) um
	42.3(2.0) %	32.9(3.4) %

The rank order of rapid elastic recovery with the tablets compressed at 30 MPa was the same as that for total elastic recovery of tablets compressed at low compressional pressures (Table 2). Rapid elastic recovery was clearly smallest for ACP tablets agreeing with the earlier discussion about the most effective consolidation and bond formation due to the fragmentation of ACP agglomerates. The difference between DCP tablets and Avicel and Emcocel tablets was much more pronounced in rapid elastic recovery than the difference in total elastic recovery (Fig. 6). Celik and Travers (22) have pointed out that slower viscoelastic expansion of tablet occuring after rapid elastic recovery is less likely to cause for example lamination

of the tablet than rapid elastic recovery. Our results agreed with this concept. The clearly loosest tablet structure (Fig. 4) of DCP tablets as well as the clearly lowest compact strength of DCP tablets observed in previous study (13) obviously resulted from the greatest rapid elastic recovery of these tablets, which in turn resulted from the inability of this material to establish firm attraction between adjacent particles after the load was removed.

The difference in rapid elastic recovery, especially between ACP tablets and Avicel and Emcocel tablets, became smaller with the tablets compressed at 90 MPa (Table 2), but the rank order between the materials remained the same as with the tablets compressed at 30 MPa and correlated with the strength of cellulose tablets (13). Total deformation under testing pin (13) as well as the absolute rapid elastic recovery were clearly smaller with the tablets compressed at 90 MPa compared to those of tablets compressed at 30 MPa.

CONCLUSIONS

Agglomerated cellulose powder (ACP) had slightly largest apparent net work values and according to yield pressures also slightly greatest tendency to both total and plastic deformation. This small difference for the advantage of agglomerated cellulose powder could be due to the most amorphous structure of this material. However, the differences in net work and in yield pressures were small between all the cellulose materials and could not alone be an explanation for the previously found clear differencies in the ability of these materials to form strong compacts. The obvious ability of the agglomerated cellulose powder to fragmentate at small compressional pressure into smaller plastically deforming particles was, on the contrary, advantageous for tablet formation.

Both total and rapid elastic recovery of tablets showed similar kind of rank order for the cellulose materials studied. The recoveries for compressed tablets were smallest for agglomerated cellulose tablets and greatest for depolymerized cellulose (DCP) tablets. The previously measured strength of cellulose tablets correlated well with the elastic recoveries of tablets. Thus the ability of cellulose particles to establish interparticle attraction affected the differences in the ability of the celluloses to form strong compacts more than the deformation of cellulose powders during compression. The extensive rapid elastic recovery was on the basis of tablet strength more harmfull than total elastic recovery of tablets.

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