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Tablet bond. I. A theoretical model

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Summary

A model is developed that describes the processes involved in tablet bonding. The processes occurring at the contact region between two particles are developed first; an average value is assumed. This is multiplied by the number of contacts in a cross-section to obtain the tensile strength. It is assumed that: (a) plastic deformation occurs during the compression; (b) the separation of true contact areas follows the principles of fracture mechanics; (c) viscoelastic properties increase the strength because of the effects on the work done during the separation of surfaces and on the radius of curvature of the surfaces in contact; and (d) in tension all points of contact are deformed sufficiently near to their maximum strength for the observed tensile strength to be close to the sum of their individual strengths. For some materials, two mechanisms are needed to describe the strength over a complete range of solid fractions. At low solid fractions, ductile extension of the contact region may occur when certain criteria, as derived herein, are met. Otherwise, the strength is determined by a brittle mechanism. The transition from the ductile to the brittle mechanism occurs when the unloaded radii of curvature of the contacting surfaces increase beyond an upper limit as a result of the plastic deformation during compression.

Introduction

The objective of this work is to apply a theory of adhesion to the interpretation of the strength of compacts, i.e. tablet bond. As much as possible, the equations are expressed in physical parameters which are relatively easy to measure or estimate. This changes the appearance of the equations without a loss of rigor. A starting point for modeling tablet bond is to first examine the processes in the contact region of two particles. The area of contact between particles formed during compression may be discussed as a region of ad/cohesion. These regions are subjected to partial separation stress as the stored elastic energy from compression is unloaded and then to tensile loading if taken to fracture by applied body stresses. A careful examination of the properties that control the strength of the interaction between the particles is needed. An initial assumption is that following the plastic deformation of compression, the recovered surfaces are essentially spherical segments whose curvature is determined by the mechanical properties of the material. The exception would be those cases where isthmus forma-

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tion occurs, i.e. ductile extension of the contact region occurs during unloading of the stored elastic energy.

Background

In an earlier paper (Hiestand, 1985) the role of plastic deformation and dispersion forces in tablet bond were explored by using a model based on inter alia reversible elastic recovery; viscoelastic effects were not considered. The conclusion was that only the weakest bonding could be explained using this model. Other studies (Johnson et al., 1971; Muller et al., 1983) were cited but not used. They analyzed the stresses in the contact regions between two spheres and indicated that the potential strength was reduced even further when the elastic modulus is low. Additional studies (Tabor, 1975; Greenwood and Johnson, 1981) have analyzed this problem and concluded that the same result can be obtained by using a fracture mechanics approach. In this case the stress concentration at the perimeter of the contact region is considered and is used to describe the conditions controlling the separation of the surfaces. In a similar manner, the adherence of viscoelastic bodies has been described (Maugis and Barquins, 1978) for various geometries and loading conditions. The energy dissipated within the crack tip as the crack propagates is an important parameter. Also the separation of surfaces has received significant attention (Chowdhury et al., 1980; Maugis and Pollock, 1984). In the general analysis, both ductile and brittle processes are considered with three controlling conditions being identified, one of which is not applicable here, viz. the condition where the contact area is less than the critical area for brittle fracture. This condition is not considered because the attraction in the at rest state, no external load, provides an area of contact larger than the critical area. Therefore, the contacts contributing to the strength of a compact have areas during compression much larger than the critical area that determines the tensile strength for the brittle mechanism. (This becomes obvious when equations developed later are considered, see discussion following Eqn 22.)

The inclusion in the theory of the effects of viscoelastic properties is of most interest for this study. The approach used herein is somewhat simplistic in that it avoids the use of the compliance function. This is done by ignoring the time history effects; thus, estimates are obtained by the substitution into linear elastic equations values determined at a specific time (Williams, 1980), e.g. the elastic modulus and the hardness. The following theoretical treatment is principally based on an approach developed by Johnson (Johnson, 1976; Greenwood and Johnson, 1981); it includes the effects of plastic deformation during the compression on the unloaded radius of curvature of the surfaces as well as the changes of the radius of curvature due to viscoelastic relaxation.

The theory of co/adhesion classically is based on an apparent work of fracture of which surface energy is only one of the components. For viscoelastic materials the energy dissipated at the tip of the crack, where separation occurs, must have a rate dependent term (Andrews and Kinloch, 1973; Kendall, 1974; Maugis and Barquins, 1978). These often are based on considerations of the velocity of the crack propagation as the surfaces separate. An effort is made herein to interpret these viscoelastic effects as apparent changes of elastic modulus (Johnson, 1976) and hardness. Thus, the surface energy is retained as an independent term as done in the Johnson approach (Johnson, 1976). This choice of approach is arbitrary but has appeal because data collected in simple procedures may then be used to estimate many of the values needed. The choice is made, faute de mieux, even though the area of contact between particles in some specific studies is not correctly described by any existing theories (Rimai et al., 1989) including the Johnson model on which this is based. However, the merits relative to tablet bond can only be assessed by using the model to compare calculated and experimental values. The first step for this is to develop and extend the Johnson model for use in this study.

To sum the interactions in a cross section of the compact, a packing factor and coordination number are included. These will be treated much as in reference 1 but with minor changes. This provides an estimate of the number of contacts that must be separated during fracture of a unit cross section. The summed forces are equated to the tensile strength of the compact. This is a 'microstructure' interpretation of tensile strength and would be true only if the strength is not determined by a 'macrostructure' defect or stress concentrator. Thus, the experimental procedures used for testing the microstructure concepts must be carefully chosen to minimize the potential that the strength is determined by macrostructure defects, in which case a continuum mechanics approach would be needed.

Studies of the adhesion of surfaces usually must consider the roughness of the surfaces. This has been included in an adhesion parameter which provides a criterion for the conditions where adhesion will be significant (Fuller and Tabor, 1975; Briggs and Briscoe, 1976). Tablet bond is adhesion but this roughness criterion need not be considered. This is true because of the extensive plastic deformation of the contacting surfaces which causes a general redistribution of the stress so that the effect of roughness is minimized, i.e. pressure is applied until roughness problems are overcome and adhesion occurs. Furthermore, the theory developed herein starts with the interaction between individual particles which might be considered to be a 'roughness' dimension. Once all the particles are being plastically deformed, many of the effects from differences among particles become abated also.

A Model for the Bond of a Tablet

Weak tablet bond

Dispersion force equations lead to the pull off force, tensile strength, for two spheres (Tabor, 1977; Hiestand, 1985) to be as in Eqn 1. This equation is retained even when the surface energy includes more than dispersion forces, i.e. the total surface energy is used. Theoretical problems with this exist, especially with solids. However, this is a traditional approach; this estimate is used herein and in many of the references cited herein. Tension is given a negative sign.

$$f_a = -2\pi R \, \Delta\gamma \tag{1}$$

where f_a is the attraction force at pull off of two spheres, R is the harmonic mean of the radii of curvature of the surfaces in contact; $R = r_1 r_2/(r_1 + r_2)$ $\Delta\gamma$ is the change of surface energy (going from interface to free surface $\Delta\gamma = \gamma_1 + \gamma_2 - \gamma_{12}$).

Throughout this article, R is the unloaded, harmonic mean radius of curvature of the contacting surfaces. If plastic deformation has occurred, the value of R will be larger than the harmonic mean of the original particle radii of the two particles (Hiestand, 1985) (also see Fig. 1).

Greenwood and Johnson (1981) have shown that the pull off force is reduced from that of Eqn 1 for many materials because of the stress concentration that occurs at the edge of the contact area. This is based on treating the separation of surfaces as a crack growth. The results were identical to those obtained by including the effects of attraction on the contact area (Johnson, 1976). This leads to Eqn 2 for the force, f, between the two attracting spheres.

$$f = \frac{4E'a^3}{3R} - \left(8\pi E'a^3 \, \Delta\gamma\right)^{1/2} \tag{2}$$

where

$$\frac{1}{E'} = \frac{1 - v_1^2}{E_1} + \frac{1 - v_2^2}{E_2}$$

E is Young's modulus and v is Poisson's ratio; the subscripts refer to the respective spheres; and a is the chordal radius of the contact region of two particles.

To obtain the pull off force, one obtains the minimum of the force in Eqn 2 as a function of the distance of separation of two spheres. The critical value of the radius associated with the minimum is obtained by setting the derivative equal to zero. Then this value of a, a_c, is substituted into Eqn 2 to obtain (Johnson et al., 1971) Eqn 3:

$$f_a' = -\tfrac{3}{2}\pi R \, \Delta\gamma \tag{3}$$

The effect of viscoelasticity on tensile strength

For viscoelastic materials the magnitude of E' is rate dependent. (The rate dependency may be described as a strain rate or simply referred to as the time scale of the process.) This feature must be incorporated into equations analogous to Eqns 2 and 3. In the earlier equations, it was assumed that the properties were not time dependent; the instantaneous value of the elastic constant was used, $E' = E_0'$. The completely relaxed modulus is the other extreme and would be the elastic constant at infinite time, E_∞'. Intermediate values will be indicated by using the t subscript, E_t'. During separation the region at the edge of the contact zone may be considered as the tip of a propagating crack. Crack propagation theory includes a theoretically infinite strain rate at the tip of the crack. Therefore, the instantaneous elastic constant, E_0', is retained in the last term of Eqn 2. However, in the first term on the right side of Eqn 2, the elastic constant acting is E_t', where the specific value depends on the rate of the process. This generalization is questionable because the processes in the vicinity of the separating surfaces may be slow enough for one to use the totally relaxed value, E_∞'. Nevertheless, the general designation is retained in this derivation. To simplify the equations, one may substitute for E_0' using $E_0' = f(t)E_t'$. Then the critical chordal radius of the contact at pull off, a_c, becomes as shown in Eqn 4 and illustrated in Fig. 1.

$$a_c^3 = \frac{9\pi R^2 f(t)\ \Delta\gamma}{8E_t'} \tag{4}$$

In the author's laboratory, the measurements used for the tableting indices (Hiestand and Smith, 1984) provide the necessary data if the time scale is controlled as needed. These are the indentation hardness, H, and the strain index, H/E', given the symbol ϵ. $f(t)$ may be expressed in terms of these parameters (Eqn 5):

$$\frac{E_0'}{E_t'} = f(t) = \frac{H_0\epsilon_t}{H_t\epsilon_0} \tag{5}$$

The subscripts refer to the time scale as with earlier terms. The hardness values used here are Meyer hardness values obtained with spherical indenters. These substitutions yield Eqn 6.

$$a_c^3 = \frac{9\pi R^2\epsilon_t^2 H_0\ \Delta\gamma}{8\epsilon_0 H_t^2} \tag{6}$$

When the critical contact radius is used to solve for the minimum pull off force, the solution is identical to the earlier case except that $f(t)$ appears again as a multiplier, to give Eqn 7. This result was obtained (Johnson, 1976) previously but with E_t' being E_∞'. This would be analogous to setting the apparent work of fracture equal to $f(t)\Delta\gamma$ for a more classical interpretation of adhesion (Andrews and Kinloch, 1973).

$$f_a'' = -\tfrac{3}{2}\pi R f(t)\ \Delta\gamma$$

$$f_a'' = -\frac{3\pi R\epsilon_t H_0\ \Delta\gamma}{2\epsilon_0 H_t} \tag{7}$$

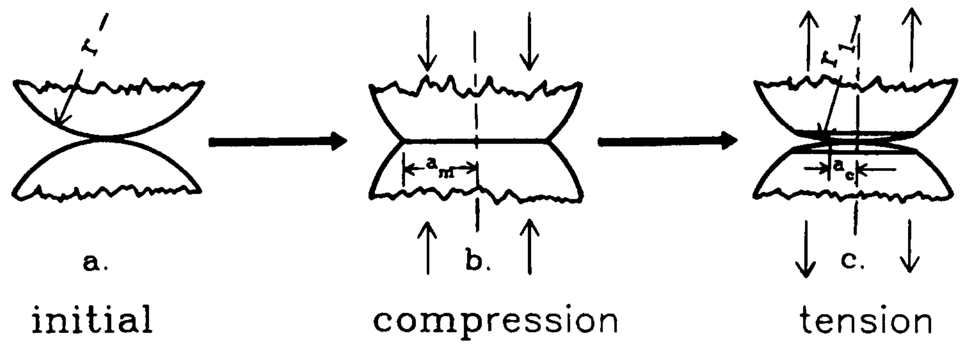

Fig. 1. (a) Initial contact uncompressed powder (contact area between particles is significant). (b) Compressed powder plastically deforms particle; contact area much larger, πa_m^2. (c) Brittle mechanism, tensile strength determined by forces when contact area is πa_c^2; Eqn 4 or 13 defines a_c.

The value of R, the harmonic mean of the radius of curvature of the unloaded surfaces in contact, is determined by the plastic deformation during compression and the influence of viscoelasticity on the elastic recovery. The recovery may be at a very different rate from the compression. Thus, R must be replaced by the appropriate terms. These will be defined later.

The effect of viscoelasticity on compression

Greenwood and Johnson (Johnson, 1976; Greenwood and Johnson, 1981) have considered the effects of viscoelasticity properties during slow compression. The force of slow compression would modify Eqn 2 by placing the $f(t)$ factor so that it appears as a divisor instead of a multiplier. The force of compression, f_c, is shown in Eqn 8; however, the terms with subscript t may refer to a different rate than for the separation process of Eqn 7.

$$f_c = \frac{4E_t' a^3}{3R} - \frac{(8\pi E_0' \,\Delta\gamma a^3)^{1/2}}{f(t)}$$

$$f_c = \frac{4E_t' a^3}{3R} - \left(\frac{8\pi E_t' \,\Delta\gamma a^3}{f(t)}\right)^{1/2} \qquad (8)$$

Since it is necessary to distinguish between the rates of the compression process and the separation process, the subscript c will be used instead of t with the respective symbols to indicate the time scale of the compression process. Eqn 8 is rewritten using these symbols plus the replacement of the E' terms to yield Eqn 9.

$$f_c = \frac{4H_c a^3}{3\epsilon_c R} - \left(\frac{8\pi\epsilon_0 a^3 \,\Delta\gamma}{H_0}\right)^{1/2}\left(\frac{H_c}{\epsilon_c}\right) \qquad (9)$$

This shows that during compression the elastic term is the dominant one; at least this is true when organic materials are used. This is stated as inequality 10 and is assumed to be true in the following discussions.

$$\frac{2a^3}{3R} \gg \left(\frac{2\pi\epsilon_0 a^3 \,\Delta\gamma}{H_0}\right)^{1/2} \qquad (10)$$

In practice, inequality 10 is used to simplify Eqn 9. Since the compression necessary to produce tablet bond must involve plastic deformation, one may set $f_c = H_c\pi a_m^2$; a_m is the maximum value of the chordal radius of contact and f_c is the force compressing the two particles together to produce a_m. (With the test compacts made for the determination of tableting indices and used for tests of these equations, the maximum compression pressure is held for 1–1.5 min.) a_m is based on the effective hardness for that time scale. The simplified Eqn 9 with f_c replaced provides the equation needed to estimate the a_m that results from the compression. Also this may be used to obtain the magnitude of R resulting from the compression, $R = 4a_m/3\pi\epsilon_c$.

Tensile strength without ductile extension

The radius of curvature of the surface can change during the time interval between compression and the testing of strength. The effective R in this experiment is influenced by all of viscoelastic stress relaxation prior to the strength test; and the radius of curvature will be larger than that acting at the time of decompression. While this cannot be known exactly, it might be reasonable to use the totally relaxed value for the strain index, ϵ_∞, instead of ϵ_c to estimate the effective value of R in a slow tensile test. In the following discussions of tensile strength, R will be estimated by using Eqn 11 which is based on the above considerations and includes both time scales, that of compression and the shelf time.

$$R = \frac{4a_m}{3\pi\epsilon_\infty} = \frac{4}{3\epsilon_\infty}\left(\frac{f_c}{\pi^3 H_c}\right)^{1/2} \qquad (11)$$

Eqn 11 is used with Eqn 7 to obtain Eqn 12 for the bond strength between two identical viscoelastic particles:

$$f_a'' = -\frac{2\epsilon_t H_0 \,\Delta\gamma}{\epsilon_\infty\epsilon_0 H_t}\left(\frac{f_c}{\pi H_c}\right)^{1/2} \qquad (12)$$

It is of interest that the tensile strength is directly proportional to a_m; and not directly proportional to the area of contact, πa_m^2, established during the compression. Using Eqn 11 in Eqn 6 yields for a_c,

$$a_c = \left(\frac{2H_0 f_c\ \Delta\gamma}{\epsilon_0 H_c}\right)^{1/3}\left(\frac{\epsilon_t}{\pi\epsilon_\infty H_t}\right)^{2/3} \quad (13)$$

or

$$\frac{a_c}{a_m} = \left(\frac{2H_0\ \Delta\gamma}{\pi\epsilon_0 a_m}\right)^{1/3}\left(\frac{\epsilon_t}{\epsilon_\infty H_t}\right)^{2/3} \quad (14)$$

Clearly, a_c/a_m decreases as the compression force increases.

Criteria for isthmus formation

Under certain conditions, the brittle mechanism that leads to the contact radius of a_c does not prevail. This is a result of the plastic deformation of the contacting surfaces during both the unloading of elastic stresses and the application of tensile stresses. This of course alters the stress distribution. Fig. 2 diagrammatically represents this process. If one assumes that ductile extension of the contact zone essentially is the mirror image of indentation and requires the same stress, H (of opposite sign), one may establish a criterion for ductile extension, i.e. ductile extension will occur when the last term of Eqn 2 exceeds the force necessary for ductile extension (Johnson, 1976). In part because of the low surface energy of organic materials, this would not be expected. However, if one includes the 'multiplier' of the viscoelastic effect, $f(t)$, the conditions might be met by organic materials, i.e. the interparticle attraction might be sufficient to cause plastic deformation. The condition for ductile extension is expressed in inequality 16 which is obtained from Eqn 2 by rearranging and using only the left and far right terms as shown below. The chordal radius used, a_d, is the maximum chordal radius of the interparticle contact at which ductile extension would occur.

$$H_t \pi a_d^2 = -f + \frac{4E_t' a_d^3}{3R} = \left(8\pi a_d^3 E_0'\ \Delta\gamma\right)^{1/2}$$

$$H_t = \left(\frac{8E_0'\ \Delta\gamma}{\pi a_d}\right)^{1/2} \qquad \text{where } a_m > a_d > a_c$$

The condition $a_d > a_c$ is necessary to avoid brittle fracture and will be incorporated in a more useful criterion. Solving for a_d, one obtains for the maximum value of a_d,

$$a_d = \frac{8H_0\ \Delta\gamma}{\pi\epsilon_0 H_t^2} \qquad a_m > a_d \quad (15)$$

The minimum value a_d may have is essentially a_c; thus, a_c should be used in the equation to establish a criterion. This plus substituting for the

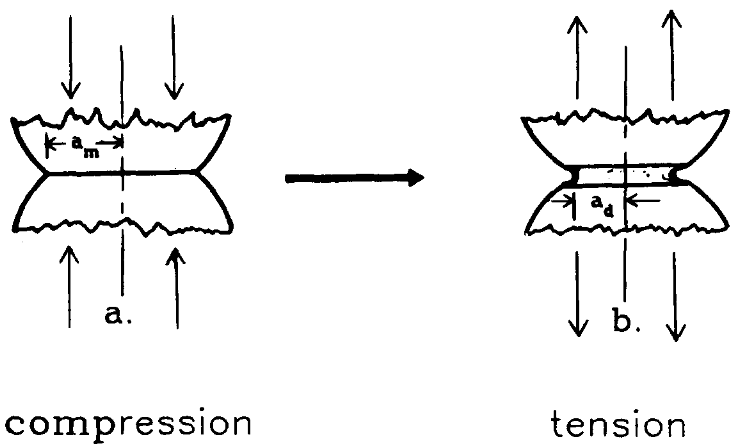

Fig. 2. (a) Contact area established during compression, πa_m^2. (b) Ductile extension during unloading of stored energy and/or tensile loading, area πa_d^2.

elastic modulus and rearranging gives inequality 16.

$$H_t < \left(\frac{8H_0\,\Delta\gamma}{\pi\epsilon_0 a_c}\right)^{1/2} \qquad a_m > a_d > a_c$$

or

$$\epsilon_0 < \frac{8H_0\,\Delta\gamma}{\pi a_c H_t^2} \qquad a_m > a_d > a_c \tag{16}$$

If 'necking' occurs during the ductile extension, the chordal radius of the contact decreases. This extension lowers the stress concentration in the region and the ductile extension would not be terminated when the contact radius reached a_c, i.e. the conditions that lead to the definition of a_c are no longer existing. Except for a_c, the quantities in inequality 16 can be experimentally estimated; a_c may be calculated for a specific compression force, f_c.

To determine if a_d is larger than a_c, which of course it must be to be controlling, one may substitute a_d for the equivalent terms into Eqn 14. After rearrangement, a simple relationship is obtained.

$$\frac{a_d}{a_c} = \left[\frac{2a_c\epsilon_\infty}{a_m\epsilon_t}\right]^2 \tag{17}$$

For ductile extension, clearly $a_d/a_c > 1$ and $a_c/a_m < 1$; therefore, $\epsilon_\infty/\epsilon_t$ must be greater than 0.5. If the material is not viscoelastic, a_c/a_m would need to be greater than 0.5 for ductile extension to occur.

A more revealing criterion for ductile extension may be obtained by setting $a_d/a_c > 1$ and using Eqn 14 to replace a_c/a_m. The compression force is inserted by replacing a_m, which was introduced with Eqn 14. After rearranging, inequality 18 is obtained:

$$f_c < \frac{256H_c}{\pi}\left[\frac{\epsilon_\infty H_0\,\Delta\gamma}{\epsilon_t\epsilon_0 H_t^2}\right]^2 \tag{18}$$

The transition from the ductile to brittle mechanism when the fully plastic loading was increased has been reported previously (Maugis and Pollock, 1984). (If the material is not viscoelastic, $f_c < (256/\pi H)\,(\Delta\gamma/\epsilon)^2$.) f_c or a_m have been introduced into the criteria through the calculation of R. While f_c and a_m could go to zero, R cannot. Therefore, one must examine the limits for the magnitude of R. The plastic deformation associated with tableting would indicate that R should be larger than the harmonic mean of the particle radii. Therefore, it is incorrect to conclude from inequality 18 that all materials will undergo ductile extension when f_c is very small. It is necessary that the particle radius be smaller than some critical value. (For irregular shape particles, it would be the radius of curvature of the contact region that must be considered.) The compression force-radius of curvature criterion becomes the most useful when discussing tablet bond. It eliminates ductile extension for many cases because the compression force is much too large for this mechanism to be controlling.

Using Eqns 6 and 15, Eqn 19 is obtained:

$$\left(\frac{a_d}{a_c}\right)^{1/2} = \frac{8a_c}{3\pi\epsilon_t R} \tag{19}$$

For $a_d/a_c > 1$, $a_c/\epsilon_t R > 1.178$. ϵ_t is expected to be less than 0.05; therefore, a_m probably must be greater than $0.0424R$ for ductile extension to occur. Later (see discussion following Eqn 22) it is shown that $a'/a_c = 4^{1/3} = 1.587$. Therefore, $a_d > 0.63a'$ for ductile extension to occur in an uncompressed powder bed. Also, multiply both sides of Eqn 19 by a_d/a_c; substituting for a_d on the right side; and setting $(a_d/a_c)^{3/2}$ to greater than one, gives inequality 20 as another criterion for ductile extension.

$$R < \frac{64H_0\,\Delta\gamma}{3\pi^2\epsilon_t\epsilon_0 H_t^2} \tag{20}$$

(If the material is not viscoelastic, $R < 64\,\Delta\gamma/3\pi^2\epsilon^2 H$.) With organic materials this could require a value of R less than that obtained from the particle radii. Furthermore, the plastic deformation resulting from compression will increase R to a value larger than the harmonic mean of the particle radii. Therefore, it is predicted that with many organic materials ductile extension is not the bonding mechanism.

Tensile strength when ductile extension occurs

If the material should meet the criterion for isthmus formation, the tensile force required to pull the particles apart would be as in Eqn 21 which assumes that ductile extension requires the same force per unit area but of opposite sign from indentation:

$$f_{\mathrm{ad}} = -H_t \pi a_d^2 = -\left(\frac{64}{\pi H_t^3}\right)\left(\frac{H_0\,\Delta\gamma}{\epsilon_0}\right)^2 \tag{21}$$

Note that if the criteria are met, the strength becomes independent of R or of a_{m}. Possibly, these equations could help with the interpretation of the cohesiveness of powders where f_{c} is very small. If the force of attraction between two particles causes the contact radius to exceed a_{c}, it is possible that ductile extension could occur during the separation if the other criteria are met.

It is interesting to compare Eqns 21 and 12. $f_{\mathrm{ad}}/f_{\mathrm{a}}''$ provides some insight relative to the strength of the adhesion between two particles and the processes occurring during the unloading, i.e. during the separation of the particles.

$$\frac{f_{\mathrm{ad}}}{f''_{\mathrm{a}}} = \frac{32\epsilon_\infty H_0\,\Delta\gamma}{\pi\epsilon_0\epsilon_t H_t^2 a_{\mathrm{m}}} \tag{22}$$

If the particles are not viscoelastic, this reduces to $f_{\mathrm{ad}}/f_{\mathrm{a}}'' = 32E'\Delta\gamma/\pi H^2 a_{\mathrm{m}}$. For most organic materials H^2 would be larger than E' perhaps by 10^4 factor. Therefore the two mechanisms can yield similar strengths.

All of the above discussion has been based on $a_{\mathrm{m}} > a_{\mathrm{c}}$ conditions. That the $a_{\mathrm{m}} < a_{\mathrm{c}}$ case considered elsewhere (Maugis and Pollock, 1984) is not a factor here can be shown by setting f equal to zero in Eqn 2 and calculating the contact radius, a', and then comparing it to a_{c} (Eqn 4). The result is $a'/a_{\mathrm{c}} = 1.6$. Therefore, in compressed compacts, the maximum area of contact always exceeds the critical contact area.

The tensile strength

Since the tablet bond strength is manifest as the strength of the compact, one must decide what property of the compact will be useful for testing the above theory. The most obvious is the tensile strength. However, structural engineers have demonstrated that the tensile strength of a large brittle body is determined by the defect structure, a defect that concentrates the stress and causes a crack to propagate (Birchall et al., 1981). Thus, the 'useable' tensile strength may be significantly less than the sum of the strength of the microstructural bonding units. The tensile strength is a macrostructural property as viewed from a continuum mechanics perspective. However, one may view a compact as being formed in a way that limits both the occurrence and the magnitude of the macrostructural defects. (Obviously pores are present. They are dispersed throughout the compact and are not considered herein to be macrostructural defects.) If a very limited volume of the compact is considered, the solid fraction gradient within the compact is very small and the variations are determined very much by the same particle properties that determine the attraction between the particles. If the compact can be made using essentially isostatic body stresses during the unloading, macrostructural defects should be avoided. Furthermore, if the method used to determine tensile strength subjects only a small volume of the compact to the maximum tensile load, the chance of the observed tensile strength being determined by a macrostructural defect would be diminished. (This departs from classical engineering notions where the important strength is that of the total structure, i.e. the weakest "link" of the chain, a macrodefect site.) At least for the theoretical case, one may calculate a tensile strength for the compact by summing the strength of the microstructural units in a cross section. This means that during a tensile test all contact points should be deformed sufficiently close to their maximum strength for the observed tensile strength to be very close to the sum of their individual strengths. It minimizes but does not eliminate the weakest link effect. Also, it depends on defining tensile strength as the fracture strength. (This differs from the engineering concept that tensile strength is the strength when plastic deformation is initiated.) To develop the above model, some estimates must be made of the total number of participating particle contacts in a cross section.

The effective number of contacts in a cross section

The number of contacts acting will be a function of solid fraction. However, the mechanism acting at the interparticle contacts also may change with solid fraction. Specifically, the criterion for ductile extension suggests that some materials may undergo isthmus formation at lower compression forces and then change to the brittle mechanism at higher compression forces. If this happens there is a transition region where some particles in the compact may be undergoing ductile extension while others are separating by crack propagation. In fact for such cases the latest points of contact established during the compression may undergo ductile extension even though most points are not. Data outside the transition region, if it exists, should be used to minimize the mixed mechanism effect. The coordination number of the particles in a compact may be dealt with as an average for a given solid fraction of a compact. Similarly the size of the contact radius at each of the contact regions is assumed to be the same, the average size. Then an estimate of the number of particles in a cross section of the tablet must be made; and an estimate of the contacts that must be separated in a fracture process can be deduced. There is a finite thickness to the 'cross section' of interest because the contact areas that would separate and therefore would contribute to the strength are not all in the same plane. It is convenient to call this a 'fracture cross section'. The assumptions used in the earlier paper (Hiestand, 1985) will be used as a starting point.

The simplest approach to the estimate of the number of particles in a cross section, N_p, would be to extend Rumpf's work to a higher solid fraction value than his studies included (Rumpf, 1962). His equation assumes a linear increase in N_p with solid fraction, ρ_r, up to a solid fraction of 0.74, $N_p = 0.4775\rho_r/r^2$ where r is the mean particle radius. In the earlier paper (Hiestand, 1985), it was assumed that the rate of increase of particles per fracture cross section relative to packing density would be at a slower than a linear rate after the close packing at ~ 0.74 solid fraction (Inadvertently a word was omitted in the text that made it read incorrectly); the solution was empirical. The assumption was that the removal of 26% of the volume in going from solid fraction of 0.74 to 1.0 would increase the number of particles in the fracture cross section by 26%. After close packing the thickness of the fracture cross section would decrease and further compression would be less effective at introducing new particles into this effective cross section. The equation used, Eqn 23, would add the desired 26% over the number at solid fraction of 0.74:

$$N_p = \frac{3(2-0.74)0.74}{2\pi(2-\rho_r)r^2} = \frac{0.445}{r^2(2-\rho_r)} \tag{23}$$

In practice, the most difficult value to assign is that of the particle radius, r. Not only the shape but the size distribution may be very broad. This model does not include these factors.

The number of points of contact in the fracture cross section must be greater than N_p because each particle is in contact with several other particles. Some literature suggests that the coordination number increases at an increasing rate after close packing has occurred (Fischmeister and Arzt, 1983). Experimental values of actual coordination numbers for spherical particles may reach 11 or 12 as the solid fraction approaches unity. Only those contacts that must be separated need be considered in tensile strength measurements. Therefore, in the earlier paper (Hiestand, 1985) the number of contacts per particle determining the strength was assumed to average three. Undoubtedly this was a very conservative estimate. If it is assumed that the number of contacts that must be separated increases linearly from three at $\rho_r = 0.74$ to four at $\rho_r = 1$, Eqn 24 would estimate the average contacts per particle in the fracture cross section, Z_e:

$$Z_e = 3.85\rho_r + 0.15 \tag{24}$$

Greater than four contacts could be involved and certainly one would not expect less than three. This too is an arbitrary correction. Studies of coordination number usually do not sort out the effective value in a fracture cross section and cannot be used directly for this problem. The empirical Eqn 25 will be used herein to estimate the number of contacts, N, that must be separated when measuring the strength of a tablet.

$$N = N_p Z_e = \frac{1.71\rho_r + 0.067}{r^2(2 - \rho_r)} \tag{25}$$

If N' is defined as equal to Nr^2, one may plot the values for the tensile strengths against N' without knowing the particle size.

Because new contacts are constantly being established during the compression, the conditions are very complex. However, if one assumes that the average f_c per particle contact in a cross section will remain essentially constant then, σ_T vs N' would be essentially linear in the absence of particle fracture during compression. Indirect support for such an assumption is that when asperity heights were exponentially distributed in a model for contact between rough surfaces, the average area of contact per asperity remained constant no matter what the load. Thus, doubling the load doubled the number of contacts (Greenwood and Williamson, 1966). The equations to be developed for the tensile strength of a compact will assume an average value for f_c can be used. If this is valid, the results will support the conclusion that the tensile strength should be a linear function of N' when N has been correctly defined. Expected exceptions may arise because particle fracture would introduce curvature and inflections could indicate a change in the bonding mechanism from ductile extension to crack propagation.

Estimating the tensile strength of a compact

The compression force acting at the interparticle interface for a given solid fraction compact will be needed in the equations for calculating the tensile strength. The experimental compression pressure, σ_c, divided by the number of contacts in a cross section, N, seems to be the most obvious estimate of this. It will be assumed that Eqn 26 is valid for the replacement of f_c:

$$Nf_c = \sigma_c \tag{26}$$

N times the interparticle force of adhesion will be used for the tensile strength of the compact, σ_T, as in Eqn 27.

$$Nf_a'' = \sigma_T \tag{27}$$

Based on the above, the uniaxial tensile strength of a compact may be estimated for the three specific cases considered. The weakest bonding case is Eqn 28 obtained by combining Eqns 3 and 11. Because the magnitude of N is independent of the bonding mechanism, it will not be replaced in the following expressions. The weakest case arises from the absence of viscoelasticity and $\epsilon_\infty = \epsilon_0$ and $H_c = H_0$. Therefore, the time indicating subscripts may be omitted for Eqn 28:

$$\sigma_T = Nf_a' = -\left(\frac{2\,\Delta\gamma}{\epsilon}\right)\left(\frac{N\sigma_c}{\pi H_c}\right)^{1/2} \tag{28}$$

Eqn 28 is just a special case of the equations for the viscoelastic case. The effects of viscoelasticity on the above model gives Eqn 29:

$$\sigma_{Tv} = Nf_a'' = -\left(\frac{2\epsilon_t H_0\,\Delta\gamma}{\epsilon_\infty \epsilon_0 H_t}\right)\left(\frac{\sigma_c N}{\pi H_c}\right)^{1/2} \tag{29}$$

If one assumes that the instantaneous values cancel, then the change in the equations when viscoelasticity is included gives the following ratio.

$$\frac{\sigma_{Tv}}{\sigma_T} = \frac{\epsilon_t H_0}{\epsilon_\infty H_t} = \frac{f(t)\epsilon_0}{\epsilon_\infty} \tag{30}$$

Eqn 30 contains the expected $f(t)$ and the effects resulting from the influence of viscoelasticity on the radius of curvature, R. This may increase the strength more than $f(t)$ alone would.

The case of isthmus formation gives Eqn 31 which is obtained from Eqns 21.

$$\sigma_{Td} = Nf_{ad} = -\left(\frac{64N}{\pi H_t^3}\right)\left(\frac{H_0\,\Delta\gamma}{\epsilon_0}\right)^2 \tag{31}$$

Before applying Eqn 31, it is necessary to establish that isthmus formation would occur. If not, Eqn 29 would be suitable since it would reduce to Eqn 28 whenever viscoelastic properties are not present.

It must be recognized that as defined in Eqn 25, N is not a continuous function covering the range from a loose powder to a compact of solid fraction equal to unity. Also, the strength of the compact would go to zero, long before N would become zero. Therefore, the equations given for strength vs N should have an intercept value. The above derivations have not provided this term. However, this is estimated by starting the σ_T plot for the same N' as that obtained by extrapolating the σ_c plot to the N' axis. However, the slopes of the equations with or without the intercept term would be the same. Therefore, it is possible to check the equations either for absolute value or for the rate of change. The latter is the subject of part II, a separate paper.

Conclusions

Equations have been derived that describe two different mechanisms that may occur during the separation of surfaces following the compression that produces plastic deformation. Criteria have been derived that establish when a ductile mechanism, isthmus formation, will occur. When these criteria are not met, a brittle mechanism occurs. With the same material, lower compression forces may be followed by the ductile process, but at higher compression forces the brittle mechanism will occur. This transition occurs because of the larger radius of curvature of the recovered surfaces. Viscoelastic properties contribute greatly to the strength of the bonding.

Glossary

Symbol	Meaning
a	chordal radius of circular contact region between spherical surfaces
a_c	critical value of a, determines pull off force; brittle mechanism
a_d	maximum chordal radius of contact for ductile extension
a_m	maximum radius of contact produced by compression
a'	chordal radius before external forces are applied
E	Young's modulus of elasticity, number subscripts refer to particle 1 and particle 2
E'	complex elastic modulus; two materials; see Eqn 2
E_0'	complex modulus for instantaneous process
E_t'	complex modulus for viscoelastic materials when the strain rate is based on the process rate using time t, $\infty > t > 0$
E_∞'	complex modulus for a relaxed viscoelastic material
f	force between two particles (compression or tension)
f_a	pull off force, separation of two spherical surfaces; equilibrium thermodynamics case
f_a'	pull off force; spherical surfaces, brittle mechanism
f_a''	pull off force; spherical surfaces; brittle mechanism, viscoelastic material
f_{ad}	pull off force; spherical surfaces, ductile extension of surfaces
f_c	compression force; spherical surfaces
$f(t)$	defined by Eqn 5, ratio of complex moduli at different strain rates
H	indentation hardness of the particle
H_0, H_t	indentation hardness, subscripts refer to associated strain rates as in E' definitions
H_c	indentation hardness of particles at strain rate used for the compression of the compact
N	the number of interparticle contacts in a cross section of the compact that must be separated during fracture
N'	Nr^2
N_p	number of particles in a cross section of the compact
r	particle radius, number subscripts refer to particle 1 and particle 2, respectively
R	the harmonic mean radius of the two contacting spherical surfaces
Z_e	mean contacts per particle separated during fracture.
γ	surface energy, number subscripts refer to particle 1 and particle 2, respectively
$\Delta\gamma$	$\gamma_1 + \gamma_1 - \gamma_{12}$ where the subscript numbers refer to particles 1 and 2 of the pair involved; the interface between them indicated by subscript 12
ϵ	H/E'; the strain index, relative strain (sphere separation) following the plastic deformation during compression
ϵ_0, ϵ_t, ϵ_c	strain indices, subscripts refer to strain rate as in definitions of E' and H
υ	Poisson's ratio, the number subscripts refer to particle 1 and particle 2, respectively
ρ_r	relative density, solid fraction, volume fraction of solid in the compact; equal to 1–porosity
σ_c	maximum compression stress on the compact when making it
σ_T	tensile strength of the compact; brittle mechanism
σ_{Tv}	compact tensile strength; brittle mechanism, viscoelastic material
σ_{Td}	compact tensile strength; ductile mechanism

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