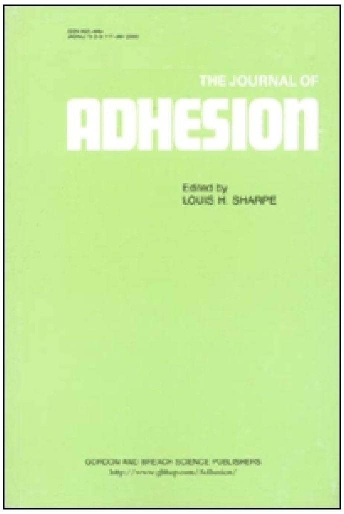

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The Effect of Pendular Moisture on the Tensile Strength of Powders

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ABSTRACT

A theory of the tensile strength of powders caused by moisture in the pendular state is derived. It also incorporates the effect of powder density and particle size distribution. It has only been possible to test the theory in a limited way (by comparison with experimental results obtained with poly-vinyl chloride powders of a given size fraction containing 6 to 17% of moisture) because published data do not include all the required information. The usefulness of the theory is, therefore, in indicating the parameters and factors that should be covered in any future experimental investigation into the effect of moisture on tensile strength.

INTRODUCTION

THE EFFECT of moisture on the tensile strength of powder compacts has been studied by a large number of workers. During the present decade, both Eisner, Fogg and Taylor[1] and Rumpf[2] determined the strength of ground limestone; Shotton and Harb[3] studied various starches, gums and sugars; Carr[4] and Richards[5] used coal powders; Fowler and Radford[6] used glass beads; and Pietsch, Hoffmann and Rumpf[7] used limestone. Recently the tensile strength of a polyvinyl chloride powder has also been measured in our Laboratory.

A more coherent situation exists with the theoretical study. One aspect of this study, common to the study of tensile strength in general, is the calculation of the number of particles in the area of split and the determination of the direction of action of the force between pairs of particles in the summation of the interparticle forces to give the tensile strength. These problems have been discussed by Rumpf and co-workers[2,8,9] and others[4,10,11] and recently by Cheng[12]. The other aspects directly relevant to the effect of moisture include the problems of distribution of moisture between pairs of particles as liquid bridges and among the voids in the compact, and of the strengths of the forces arising out of the distributed moisture. These problems have been studied by Rumpf and co-workers[2,7,9], Clark and Mason[13,14] and others[15,16,17].

However, most of the theoretical work done to date has been limited to compacts of smooth equal spheres so that in the comparison with experimental results a mean equivalent particle size has to be invoked. The effect of particle size on particle distribution has been considered by Turba and Rumpf[8] and by Cheng[12], but the properties and forces due to liquid bridges between unequal particles has only been little studied (see references quoted by Clark and Mason[13]). The complication caused by rough particles has also been by-passed[7,9].

Despite the advances already made in the theoretical study of the effect of moisture on tensile strength, it is clear that the rigorous approach adopted by previous workers is bound to be impeded because of the direct and indirect effect of various factors, notably the following: (i) non-spherical and rough particles, (ii) unequal particle sizes, and (iii) unequal distribution of moisture both between pairs of particles of given sizes and between differently sized groups of pairs of particles. In the present paper a different approach is used to treat the experimental results: the method is based on the investigation developed for dry powders[12]. In this method, instead of summing theoretically derived interparticle forces to calculate the tensile strength, the experimental data is used to deduce an average value of the interparticle force. This can then be compared with the theoretical interparticle force if required, but the practical use of the derived interparticle force would be for the correlation of experimental data and thus the prediction of tensile strengths.

DERIVATION OF THEORY

The physical basis of the theory has been described in a previous paper[12]. Briefly the tensile strength, i.e. the force per unit area required to split a powder compact, is obtained by summing the contributions arising from the interparticle forces acting between pairs of particles that lie "in" the plane of the split or failure.

A powder compact consists of a collection of individual solid particles of various sizes arranged in a random lattice. Each particle is surrounded by a number, the co-ordination number, of particles that are its nearest neighbours. Such pairs of nearest neighbours will be termed "particle-pairs". The co-ordination number is known to depend on the density of the compact and from the data quoted by Rumpf[2] and Clark and Mason[13] for equal spheres, it is seen that they are related by the equation:

$$c = k/\left(1 - \frac{\rho_p}{\rho_s}\right) \tag{1}$$

where k is of the order of 3. In the present work, it will be assumed that Eq. (1) holds for rough, non-spherical and unequal particles also, except that k will vary with roughness, non-sphericity and particle size distribution.

When a powder compact is split, certain particle-pairs separate, leading to a rugged surface as the "plane" of failure which is normal to the direction of the tensile force. The separated particle-pairs will be referred to as being in the plane of failure and their number may be calculated as follows. If two surfaces are drawn one on either side of the failure plane, to pass through the centres of the individual particles of the split particle-pairs, they will enclose a volume, the volume of split, per unit area of the failure plane:

$$v = \bar{d} \tag{2}$$

where

$$\bar{d} = \sum_{i \leq j} x_{ij} \tfrac{1}{2} \left[\tfrac{1}{2}(d_i + d_{i+1}) + \tfrac{1}{2}(d_j + d_{j+1}) \right] \tag{3}$$

(It is implied that the particle size distribution is given as fractions, n_i, of total number of particles between size (equivalent spherical diameter) d_i and d_{i+1}, so that the chance of finding a pair of particles, one of diameter in the range d_i to d_{i+1} and the other d_j to d_{j+1}, is

$$x_{ij} = n_i n_j \Big/ \sum_{i \leq j} (n_i n_j) \tag{4}$$

The convention that $d_i \leq d_j$ when $i \leq j$ is used). The concentration of particle-pairs in the volume of split would be the same as in the bulk of the compact. There the number of particle-pairs per unit volume is

$$N_{pp/v} = \tfrac{1}{2} c N_{p/v} \tag{5}$$

where the factor of ½ allows for the fact that each particle-pair involves two particles. The number of particle-pairs per unit area of the failure plane is obtained from Eq. (1), (2) and (5).

$$\begin{aligned} N_{pp/a} &= N_{pp/v}\, v \\ &= \tfrac{1}{2} \frac{k}{p} N_{p/v}\, \bar{d} \\ & \quad 1 - \frac{\rho_p}{\rho_s} \end{aligned} \tag{6}$$

The number of particles per unit volume can be related to the particle size distribution and the density of the powder compact using a mass balance:

$$N_{p/v} = \frac{\rho_p}{\rho_s} \Big/ V \tag{7}$$

where

$$\rho_p = (1 - w)\, \rho \tag{8}$$

and

$$V = \sum_i \frac{\pi}{6} n_i \tfrac{1}{2} (d_i^3 + d_{i+1}^3) \tag{9}$$

Thus Eq. (6) may be rewritten in terms of experimental quantities:

$$N_{pp/a} = \frac{k}{2} \frac{\rho_p/\rho_s}{1-(\rho_p/\rho_s)} \frac{\bar{d}}{\bar{v}} \tag{10}$$

Turning now to the interparticle force arising from the presence of moisture, it is seen that moisture can exist in a powder compact in three forms depending on the amount present. At less than saturation, water molecules are adsorbed on the surface of the solid particles and no free water is present. The adsorbed molecule affects the strength of the electro-chemical or van der Waal's forces acting between the particles. The treatment of this kind of forces has been considerable in the earlier paper[12]. As the free water content increases, liquid bridges form between pairs of particles (pendular state) and the interparticle forces become dominated by surface tension and capillary forces, which are stronger than van der Waal's forces and act over longer distances. The theory of the present paper will be concerned with this type of forces. With increased moisture content, the transition or funicular state will first be reached, in which three or more particles become bound by the same "drop" of water. Finally, when all the voids are occupied, the capillary state is reached. Tensile strengths of powders in these states have been studied by Newitt and Conway-Jones[18] and by Rumpf and co-workers[2,7] and will not be considered in the present paper.

Before the interparticle force between pairs of particles is considered, it is noted that, in general, the action of this force will be at an angle, θ, to the direction of the tensile force, and it thus has an effective component proportional to cos θ. If it is assumed that the angle is uniformly distributed in the half-space, the mean effective component of the interparticle force, F, is then,

$$\bar{F} = \tfrac{1}{2}F \tag{11}$$

The interparticle force due to liquid bridges between uniform spheres has been studied by Clark and Mason[14] and Pietsch and Rumpf[9]. Both groups of workers show that the interparticle force follows the expression approximately:

$$F = \pi d \gamma f_o \left(1- \frac{t}{t_o}\right) \tag{12}$$

In Eq. (12), f_o and t_o depend on the volume of the liquid bridge and the contact angle between the liquid and the solid. Some results for $\delta = 0$ are shown in Fig. 1, from which it is seen that the dependence of f_o and t_o on volume may be expressed approximately by power law expressions:

$$f_o = 1/V^{0.045} \tag{13}$$

$$t_o = mV^{0.4} \tag{14}$$

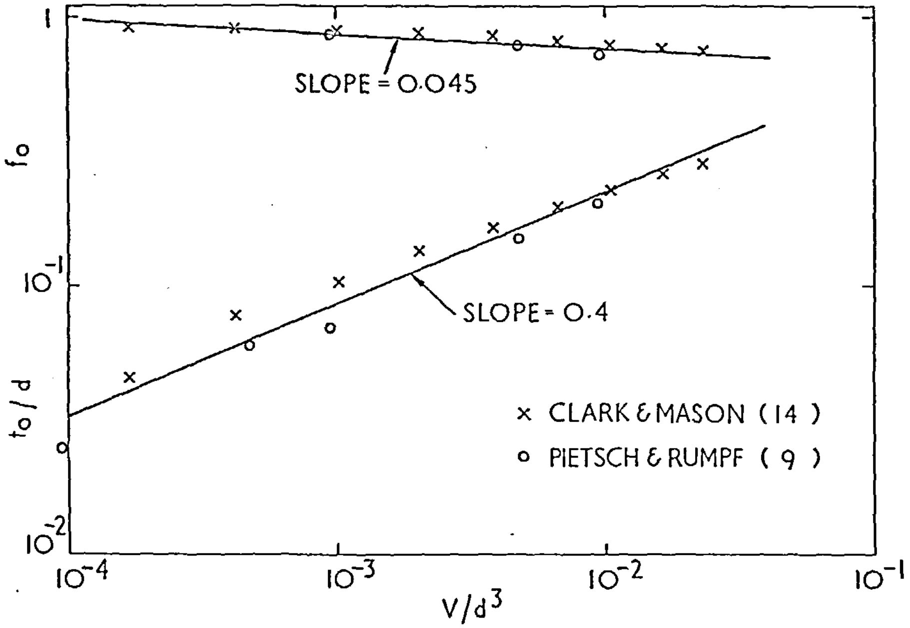

Figure 1. Dependence of f_o and t_o on the liquid bridge volume for equal spheres with zero contact angle.

The difficulty with actual powders, arising because of the causes mentioned in the introduction, makes it impossible to derive theoretical expressions for interparticle forces. To overcome this, a different approach is used in this paper. From the form of Eq. (12), (13), and (14), it will be assumed that the interparticle force between any kind of particles is given by:

$$F = \chi_{ij}\,\Psi_o\, g\left(\frac{t}{t_o}\right) \tag{15}$$

In this expression, the interparticle force has been separated into various factors: one, χ_{ij}, depending on the particle size of the particles in the particle-pairs; the other, Ψ_o, depending primarily on the liquid bridge volume and may also involve particle surface geometry and thus in turn, particle size again; and the third, $g(t/t_o)$, depending on the particle separation. By analogy with Eq. (13), it is assumed that

$$\Psi_o = L/V^{0.045} \tag{16A}$$

It is assumed that $g(t/t_o)$ is a monotonically decreasing function such that when $t = 0$, $g = 1$ and when $t = t_o$, $g = 0$. Thus, F falls to zero at some separation, t_o, which is assumed to depend on moisture content by an equation similar to Eq. (14):

$$t_o = MV^{0.4} \tag{16B}$$

In trying to sum up the contributions from all the liquid bridges in the failure plane to give the tensile strength, an uncertainty arises regarding the distribution of moisture between the particle-pairs. Pietsch et al.[7] assumed that the liquid "is uniformly distributed at all co-ordinate points" and Clark and Mason[14], while allowing for certain particle-pairs to be unbridged, assumed all liquid bridges are of the same volume. It is clear that in general particle-pairs of the same sizes may have bridges of different volumes and the proportions of particle-pairs of any size group that are bridges may differ from group to group. However, if it is assumed that there is a most likely distribution of liquid amongst the particle-pairs, then two average qualities may be considered. The first is the average bridge volume to be used in place of V and the second is the fraction of particle-pairs in any size group that are bridged. It is assumed that both of these are proportional to moisture content. Thus:

$$\Psi_o = \lambda / w^{0.045} \tag{17}$$

$$t_o = \mu w^{0.4} \tag{18}$$

$$y_{ij}(d_i, d_j, w) = y_o(d_i, d_j) w \tag{19}$$

Remembering that the proportion of particle-pairs of sizes d_i and d_j among all particle-pairs is x_{ij}, the tensile strength may now be obtained from the equation:

$$T = \sum_{i \leq j}\sum y_{ij}\, x_{ij}\, N_{pp/a} \tfrac{1}{2} F \tag{20}$$

in which Eq. (11) has been invoked. Using Eq. (10), (15), (17) and (19):

$$T = \phi\, w^{0.955} \frac{\rho_p/\rho_s}{1-(\rho_p/\rho_s)}\, g\left(\frac{t}{t_o}\right) \tag{21A}$$

$$\phi = \frac{y}{4}\left[\sum_{i \leq j} x_{ij}\, k\, y_o\, \lambda\, \chi_{ij}\right]\frac{\overline{d}}{\overline{v}} \tag{21B}$$

To complete the theory, the particle separation is related to the compact density by the method of the previous paper[12]:

$$\frac{\overline{d} + t}{\overline{d} + t_o} = \left(\frac{\rho_{po}}{\rho_p}\right)^{1/3} \tag{22}$$

which leads to the expression for the separation:

$$t = \left(\frac{\rho_{po}}{\rho_p}\right)^{1/3} \mu w^{0.4} - \left[1 - \left(\frac{\rho_{po}}{\rho_p}\right)^{1/3}\right]\overline{d} \tag{23}$$

in which Eq. (18) has been incorporated.

COMPARISON WITH EXPERIMENTAL DATA

Very little of the experimental data that exists contains all the relevant information for treatment by the theory derived in this paper. The theory can be compared with limited results obtained with a polyvinyl chloride powder. The particle density was 1.1 g/cm^3 and the particle size distribution, measured with a Coulter Counter, lies between 30 and 200 microns with a median of 110 microns. The tensile strengths were measured using the improved apparatus described by Ashton et al.[19] and the moisture contents were determined using a micro-wave Automatic Attenuation Meter (Microwave Instruments Ltd., North Shields, England). The results of tensile strength as a function of compact density for a range of moisture contents are shown in Fig. 2.

The treatment of experimental data deals with the problem of the determination of the functional form of g(t/t$_o$) and the numerical values of the other parameters. Rewriting Eq. (21A) and (23)

$$\phi\, g\left(\frac{\rho_{po}}{\rho_p}\right) = T \Big/ \left[w^{0.955} \frac{\rho_p/\rho_s}{1-(\rho_p/\rho_s} \right] \tag{24}$$

$$\frac{t}{\bar{d}} = \left(\frac{\rho_{po}}{\rho_p}\right)^{1/3} \left(w^{0.4} \frac{\mu}{\bar{d}} + 1 \right) - 1 \tag{25}$$

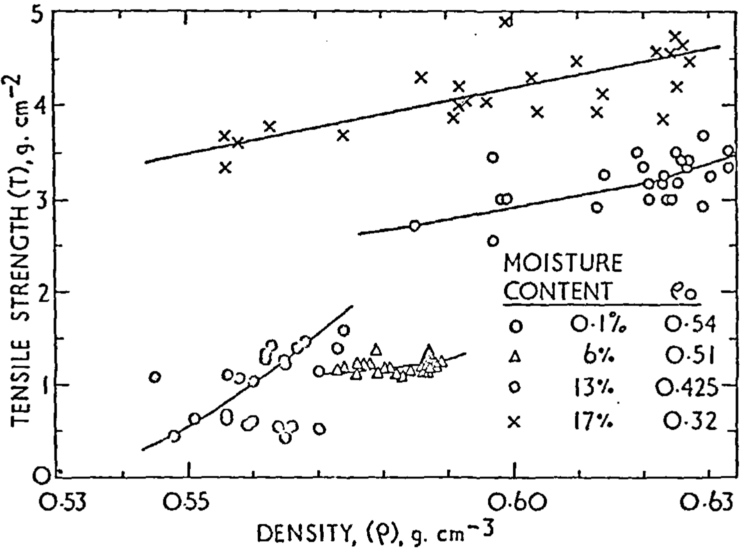

Figure 2. Experimental results of tensile strength for a polyvinyl chloride powder.

From these equations the functional form of g $\left(\frac{t}{t_0}\right)$ may be obtained from plots of the RHS of Eq. (24) versus t/t_0 using different trial values of $\mu/\bar{d}$. In calculating t, values of ρ_0 have been obtained from Fig. 2 by extrapolating the data to zero T. It was found that the data for $w = 0.1\%$ gave values of $\phi g(t/t_0)$ vastly in excess of the values obtained for the other sets of data. It also seems obvious from the small moisture content that the interparticle force involved cannot be due to liquid bridges. This set of data has therefore been excluded in the subsequent treatment of the remaining three sets of results. Because of the scatter of the data, which is considered to be unavoidable in powders, it was found that apparently good correlations between them may be obtained with different values of assumed $\mu/\bar{d}$ (Fig. 3). In Fig. 3, the results for three values of $\mu/\bar{d}$ are shown. The three fitted curves are shown, but only the data points for $\mu/\bar{d} = 0.5$ have been included.

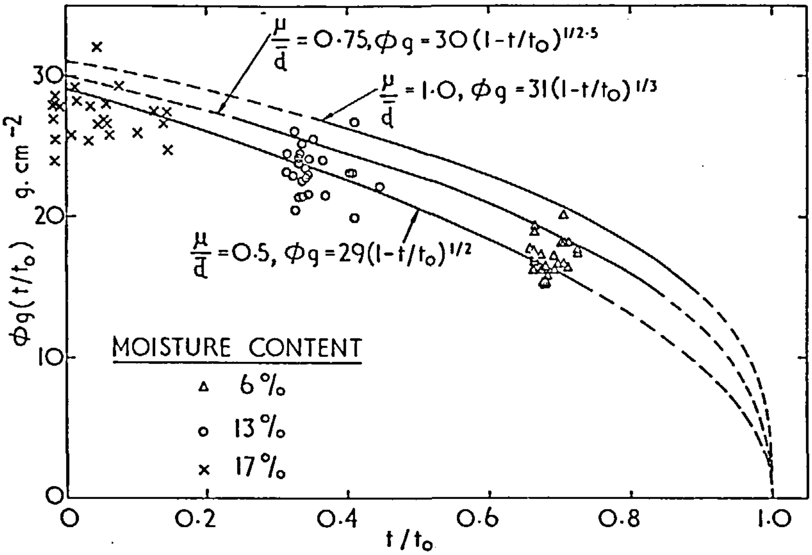

Figure 3. Correlation of tensile strength data of Figure 1 showing the dependence of interparticle force on particle separation.

The choice of the best value of $\mu/\bar{d}$ may be made by the following argument. The larger of $\mu/\bar{d}$ may be rejected because they give rise to values of $t_0/\bar{d}$ that are too large, by reference to Fig. 1, and indicate that the associated bridge volumes would be so great that the moisture would be found in the funicular state. This is clearly not so, because the critical moisture content for this to happen has been estimated, using the information of Pietsch et al.[7], to be about 17% for the highest density involved when $t = 0$. In addition, it is seen from Fig. 3, that using the value $\mu/\bar{d} = 0.5$, the experimental data for $w = 0.17$ at the high densities give values of t that are very nearly zero and thus fit well with the details of the calculation of the critical moisture content. It can therefore be concluded that the best value of $\mu/\bar{d}$ is in fact nearly 0.5 and that, instead of the linear relation of Eq. (12), an approximate square root relation is found for the dependence of interparticle force due to liquid bridges in the actual polymer powder used:

$$\phi g\left(\frac{t}{t_o}\right) = 29\left(1 - \frac{t}{t_o}\right)^{1/2} \tag{26}$$

where the unit of the numerical parameter is g/cm^2 and is valid when w is expressed as weight fraction.

DISCUSSION AND CONCLUSION

The theory of the effect of moisture, when present in the pendular state, on the tensile strength of powders differs from previous work in three respects. The first is that, in the derivation, an allowance has been made to include the effect of particle size distribution, as it affects the numbers of particle-pairs composed of different sized particles. The second is an attempt to cast the theory in such a form that the functional form of the dependence of interparticle force on particle separation due to moisture may be deduced from experimental data. This requires the third innovation, namely the calculation of an averaged particle separation from experimental quantities.

The effects of particle size distribution and moisture are, of course, intertwined, but in the theory, it has been possible to separate them. This is done in Eq. (15) from an analogy with the behaviour of liquid bridges between uniform spheres and also in Eq. (17), (18) and (19). Because the existing experimental data do not allow the effect of particle size distribution to be tested, the dependence of the various parameters that are affected by this factor, namely k, y_o, λ, χ_{ij} and μ, has not been discussed in detail above. However, because of the separation of the variables, it has been possible to test the theory and assess the effect of moisture in isolation. The result of this is shown in Fig. 3.

A further assumption is implied in the derivation of Eq. (22), namely, that the powder mass is rigid and suffers no appreciable tensile deformation up to the point of rupture. While this is substantially valid for dry powders, it is not true for powders containing free moisture. However, until the stress-strain relationship of moisture powder masses has been determined, it would be difficult to improve on Eq. (22).

While some degree of support for the theory is obtained from the experimental data, it cannot be considered that the indication of the theory is at all firm, because of the paucity of the data both in respect of the range of t covered at each w and in respect of particle size distribution. The way to judge the theory at present is not whether it has or has not the support of experimental results but through its usefulness in indicating the parameters and factors that have to be covered in any future experimental investigation into the effect of pendular moisture on tensile strength.

NOMENCLATURE

c	co-ordination number
d	particle diameter
$\bar{d}$	mean effective diameter of particle
f_o	function giving the effect of liquid bridge volume and contact angle on interparticle force between uniform spheres
F	interparticle force
$\bar{F}$	average of interparticle force over all orientations
$g(t/t_o)$	function giving the effect of particle separation on interparticle force
i, j	integers
k	parameter in Eq. (1)
l	parameter in Eq. (13)
L	parameter in Eq. (16A)
m	parameter in Eq. (14)
M	parameter in Eq. (16B)
n_i, n_j	fractional number of particles in size fraction
$N_{p/v}$	number of particles per unit volume
$N_{pp/a}$	number of particle-pairs per unit area
$N_{pp/v}$	number of particle-pairs per unit volume
t	particle separation
T	tensile strength
$\bar{v}$	mean volume of particle
v	volume of split per unit area of macroscopic split
V	volume of liquid bridge
w	moisture content by weight
x_{ij}	fraction of particle-pairs consisting of particles of sizes d_i and d_j
y_{ij}	fraction of particle-pairs of sizes d_i and d_j that is connected by liquid bridges
y_o	parameter in Eq. (19)
γ	surface tension
δ	contact angle
λ	parameter in Eq. (17)
μ	parameter in Eq. (18)
ρ	density
ρ_p	total weight of particles per unit volume of powder
ρ_s	density of particles
ϕ	defined by Eq. (21B)
χ_{ij}	function in Eq. (15)
Ψ_o	function in Eq. (15)

subscripts

i,j pertaining to particles of diameters d_i, d_j

0 pertaining to conditions at which the tensile strength vanishes

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