FLOW PROPERTIES OF COHESIVE NANOPOWDERS

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Abstract
The fundamentals of cohesive powder consolidation and flow behaviour using a reasonable combination of particle and continuum mechanics are explained. By means of the model “stiff particles with soft contacts” the influence of elastic-plastic repulsion in particle contacts is demonstrated. With this as the physical basis, the stationary yield locus, instantaneous yield loci and consolidation loci, flow function and compression function are presented. The flow properties of a very cohesive titania nanopowder \( (d_s = 200 \text{ nm}) \) are shown. These models are used to evaluate shear cell test results as constitutive functions for computer aided apparatus design for reliable powder flow.

Keywords: Particle mechanics, adhesion force, van der Waals forces, constitutive models, cohesive powder, flow properties
INTRODUCTION

The well-known flow problems of cohesive particulate solids in storage and transportation containers, conveyors or process apparatuses - mainly mentioned by Jenike (1964) - leads to bridging, channelling, oscillating mass flow rates and particle characteristics associated with feeding and dosing problems. Taking into account this list of selected technical problems and hazards, it is worth to deal with the fundamentals of particulate solids consolidation and flow, i.e., to develop a reasonable combination of particle and continuum mechanics. This method appears to be appropriate to derive constitutive functions on physical basis in the context of micro-macro transition of particle-powder behaviour.

PARTICLE CONTACT CONSTITUTIVE MODEL

The well-known failure hypotheses of Tresca, Coulomb-Mohr, the yield locus concept of Jenike (1964) and Schwedes (1988) were supplemented from Molerus (1975, 1978) by the cohesive steady-state flow criterion. The consolidation and non-rapid, frictional flow of fine and cohesive particulate solids was explained by the adhesion forces at particle contacts. His advanced theory is the suitable basis of the extended model approach, Tomas (1991, 2001, 2001a) which is shown now.

In principle, there are four essential mechanical deformation effects in particle-surface contacts and their force-response behaviour can be explained as follows:

1. **elastic** contact deformation (Hertz (1882), Huber (1904), Mindlin (1953), Dahneke (1972), Derjagin (DMT theory) (1975), Johnson (JKR theory) (1985), Thornton (1991) and Sadd (1993)) which is reversible, independent of deformation rate and consolidation time effects and valid for all particulate solids;

2. **plastic** contact deformation with adhesion (Krupp (1967), Molerus (1975, 1978), Schubert (1976), Maugis (1984), Walton (1986) and Thornton (1998)) which is irreversible, deformation rate and consolidation time independent, e.g. mineral powders;
(3) **viscoelastic** contact deformation (Yang (1966), Rumpf (1976) and Sadd (1993)) which is reversible and dependent on deformation rate and consolidation time, e.g. soft particles as bio-cells;

(4) **viscoplastic** contact deformation (Rumpf (1976)) which is irreversible and dependent on deformation rate and consolidation time, e.g. nanoparticles fusion.

These force-displacement models are shown as characteristic constitutive functions in Fig. 1. Based on these theories, a general approach for the time and deformation rate dependent and combined viscoelastic, elastic-plastic, viscoplastic, adhesion and dissipative behaviours of a spherical particle contact was derived, Tomas (2000, 2001), and is briefly explained here - a comprehensive review deals with all derivations in detail, Tomas (2003):

During approaching of the spheres, the adhesion force $F_{\text{H0}}$ without any additional contact deformation, see Fig. 2a), can be modelled as a single rough sphere-sphere-contact, additionally, with a characteristic hemispherical micro-roughness height or radius $h_r < d$ instead of particle size $d$, Schubert (1982) and Rabinovich (2000):

$$F_{\text{H0}} = \frac{C_{\text{H,abs}} \cdot h_r}{12 \cdot a_{p=0}^2} \left[ 1 + \frac{d / h_r}{2 \cdot (1 + h_r / a_{p=0})^2} \right] \approx \frac{C_{\text{H,abs}} \cdot h_r}{12 \cdot a_{p=0}^2}$$

If an external compressive normal force $F_N$ acts at a **soft contact** of two isotropic, **stiff**, linear elastic, mono-disperse spherical particles the previous contact point is deformed to a small contact area, Fig. 2, and the adhesion force between these two particles is increasing, Krupp (1967), Rumpf et al. (1976) and Molerus (1975, 1978).

After this loading and elastic deformation, Fig. 2b), the contact starts at $p_{\text{max}} = p_t$ with plastic yielding. This elastic-plastic contact deformation response in Fig. 2c) is given by the particle contact force equilibrium between attraction (-) and elastic as well as **soft** plastic repulsion (+) or force response ($r_k^*$ coordinate of annular elastic contact area):

$$\sum F = 0 = -F_{\text{H0}} - p_{\text{vdw}} \cdot \pi \cdot r_k^2 - F_N + p_t \cdot \pi \cdot r_{k,pl}^2 + 2 \cdot \pi \cdot \int_{r_{el}}^{r_k} p_{el}(r_k^*) \cdot r_k^* \, dr_k^*$$

Superposition provided, this leads to a very useful linear force displacement model (for $\kappa_A \approx \text{const.}$) with the particle centre approach of both particles $h_K$, shown in Fig. 3 as elastic-plastic boundary (averaged radius of particle 1 and 2 $r_{i,2} = (1/r_i + 1/r_2)^{-1}$), Tomas (2000):
\[ F_{x} + F_{h0} = \pi \cdot r_{1,2} \cdot p_{f} \cdot (\kappa_{A} - \kappa_{p}) \cdot h_{K} \]  

(3)

Thus, the contact stiffness decreases with smaller size \( d = 4 \cdot r_{1,2} \) (or micro-roughness radius) of cohesive powders and nanoparticles, predominant plastic yielding behaviour provided:

\[ k_{N,pl} = \frac{dF_{N}}{dh_{K}} = \pi \cdot r_{1,2} \cdot p_{f} \cdot (\kappa_{A} - \kappa_{p}) \]  

(4)

The plastic repulsion coefficient \( \kappa_{p} \) describes a dimensionless ratio of attractive Van der Waals pressure \( p_{VdW} \) to repulsive particle micro-hardness \( p_{f} \) for a plate-plate model:

\[ \kappa_{p} = \frac{p_{VdW}}{p_{f}} = \frac{C_{H,als}}{6 \cdot \pi \cdot a_{F=0} \cdot p_{f}} \]  

(5)

The characteristic adhesion distance in Eqs. (1) and (5) lies in a molecular scale \( a = a_{F=0} \approx 0.3 - 0.4 \text{ nm} \). It depends mainly on the properties of liquid-equivalent packed adsorbed layers and can be estimated for a molecular interaction potential minimum \( -dU/da = F = 0 = F_{at} + F_{rep} \) or force equilibrium. Provided that these molecular contacts are stiff enough compared with the soft particle contact behaviour influenced by these mobile adsorption layers due to molecular rearrangement, this separation \( a_{F=0} \) is assumed to be constant during loading and unloading in the interesting macroscopic pressure range of \( \sigma = 1 - 100 \text{ kPa} \). The Hamaker constant solid-liquid-solid \( C_{H,als} \) due to Lifschitz theory is related to continuous media and depends on their dielectric constants and refractive indices, Israelachvili (1992).

The elastic-plastic contact area coefficient \( \kappa_{A} \) represents the ratio of plastic particle contact deformation area \( A_{pl} \) to total contact deformation area \( A_{K} = A_{pl} + A_{el} \) including a certain elastic displacement, Tomas (2000)

\[ \kappa_{A} = \frac{2}{3} + \frac{1}{3} \frac{A_{pl}}{A_{K}} = 1 - \frac{1}{3} \sqrt{\frac{h_{K,f}}{h_{K}}} \]  

(6)

with the centre approach \( h_{K,f} \) for incipient yielding at point Y in Fig. 3, \( p_{el}(r_{K} = 0) = p_{max} = p_{f} \)

\[ h_{K,f} = d \left( \frac{\pi \cdot p_{f}}{2 \cdot E^{*}} \right)^{2} \]  

(7)

and the averaged modulus of elasticity \( E^{*} \) of both particles 1 and 2 (\( \nu \) Poisson’s ratio).
\[ E^* = 2 \left( \frac{1 - \nu_1^2}{E_1} + \frac{1 - \nu_2^2}{E_2} \right)^{-1} \]  

(8)

Constant mechanical bulk properties provided, the finer the particles the smaller is again the yield point \( h_{K,f} \) which is shifted towards zero centre approach. Thus, an initial pure elastic contact deformation \( A_{pl} = 0, \kappa_A = 2/3 \), has no relevance for cohesive nanoparticles and should be excluded. But after unloading, beginning at point U with curve U – E, Fig. 3, the contact recovers elastically in the compression mode and remains with a perfect plastic displacement \( h_{K,E} \). For this pure plastic contact deformation \( A_{el} = 0 \) and \( A_K = A_{pl}, \kappa_A = 1 \) is obtained.

Below point E left the tension mode begins. Between U – E – A the contact recovers probably elastically along a supplemented Hertzian parabolic curvature up to displacement \( h_{K,A} \):

\[ F_{N,unload} = \frac{2}{3} E^* \sqrt{r_{1,2} \cdot \left( h_K - h_{K,A} \right)^3} - F_{H,A} \]  

(9)

But along the symmetric curve A - U the contact may be reloaded:

\[ F_{N,reload} = -\frac{2}{3} E^* \sqrt{r_{1,2} \cdot \left( h_{K,U} - h_K \right)^3} + F_{N,U} \]  

(10)

If one applies a certain pull-off force \( F_{N,Z} = -F_{H,A} \), here negative,

\[ F_{H,A} = F_{H0} + \pi \cdot r_{1,2} \cdot \rho_{vdW} \cdot h_{K,A} \]  

(11)

the adhesion (failure) boundary at point A is reached and the contact plates are failing and detaching with the increasing distance \( a = a_{vdw} + h_{K,A} - h_K \). This actual particle separation can be considered for the calculation by means of a long-range hyperbolic adhesion force curve \( F_{N,Z} = -F_{H,A} \propto a^{-3} \) with the plate-plate model as given in Eq. (5).

Additionally, if one considers a single elastic-plastic particle contact as a conservative mechanical system without heat dissipation, the energy absorption equals the lens-shaped area between both unloading and reloading curves A - U in Fig. 3:

\[ W_{disc} = \int_{h_{k,A}}^{h_{k,U}} F_{N,reload}(h_K) \, dh_K - \int_{h_{k,A}}^{h_{k,U}} F_{N,unload}(h_K) \, dh_K \]  

(12)
With Eqs. (9) and (11) for $F_{H,A}$ and Eqs. (3) and (10) for $F_{N,U}$, one obtains finally the specific or mass related energy absorption $W_{m,\text{diss}} = k \cdot W_{\text{diss}} / m_p$, which includes the averaged particle mass $m_p = 4/3 \cdot \pi \cdot r_{i,2}^3 \cdot \rho_s$. In addition, the resultant Eq. (13) includes a characteristic contact number in the bulk powder (coordination number $k \approx \pi/\varepsilon$, Molerus (1975)):

$$W_{m,\text{diss}} = \frac{-E^*}{20 \cdot \varepsilon \cdot \rho_s} \left( \frac{h_{K,U} - h_{K,A}}{r_{i,2}} \right)^{5/2} + \frac{3 \pi \cdot \rho_s \cdot \left( h_{K,U} - h_{K,A} \right)}{32 \cdot r_{i,2}^2 \cdot \varepsilon \cdot \rho_s} \left[ k_A \cdot h_{K,U} - k_p \cdot \left( h_{K,U} - h_{K,A} \right) \right]$$

(13)

This specific energy of 3 to 85 $\mu$J/g for the titania powder example mentioned was dissipated during one unloading - reloading - cycle in the bulk powder with an average pressure of only $\sigma_{M,\text{st}} = 2$ to 18 kPa (or major principal stress $\sigma_1 = 4$ to 33 kPa).

The slopes of elastic-plastic yield and adhesion boundaries in Fig. 3 are characteristics of irreversible particle contact stiffness or compliance. Consequently, if one eliminates the deformation $h_k$ the linear adhesion-normal force function $F_H = f(F_N)$ in Fig. 4 is obtained, Tomas (2000).

$$F_H = \frac{k_A}{k_A - k_p} \cdot F_{H0} + \frac{k_p}{k_A - k_p} \cdot F_N = (1 + \kappa) \cdot F_{H0} + \kappa \cdot F_N$$

(14)

The dimensionless elastic-plastic contact consolidation coefficient (strain characteristic) $\kappa$ is given by the slope of adhesion force $F_H$ influenced by predominant plastic contact failure.

$$\kappa = \frac{k_p}{k_A - k_p}$$

(15)

This elastic-plastic contact consolidation coefficient $\kappa$ is a measure of irreversible particle contact stiffness or softness as well. A shallow slope implies low adhesion level $F_H \approx F_{H0}$ because of stiff particle contacts, but a large slope means soft contacts, or i.e., a cohesive powder flow behaviour. This model considers, additionally, the flattening of soft particle contacts caused by the adhesion force $\kappa \cdot F_{H0}$. Thus, the total adhesion force consists of a stiff contribution $F_{H0}$ and a contact strain influenced component $\kappa \cdot (F_{H0} + F_N)$, Fig. 4.

This Eq. (14) can be interpreted as a general linear particle contact constitutive model, i.e. linear in forces, but non-linear concerning material characteristics. The intersection of function (14)
with abscissa ($F_H = 0$) in the negative extension range of consolidation force $F_N$ is surprisingly
independent of the Hamaker constant $C_{H,sls}$, Fig. 4:

$$
F_{N,Z} = \frac{\pi}{2} \cdot a_{F=0} \cdot h_r \cdot p_f \cdot \left( \frac{2}{3} + \frac{A_{pl}}{3 \cdot A_K} \cdot \left[ 1 + \frac{d/h_r}{2 \cdot (1 + h_r / a_{F=0})^2} \right] \right) \approx \frac{\pi}{2} \cdot a_{F=0} \cdot h_r \cdot p_f \tag{16}
$$

Considering the model prerequisites for cohesive powders, this minimum normal (tensile) force
limit $F_{N,Z}$ combines the opposite influences of a particle stiffness, micro-yield strength $p_f \approx 3 \cdot \sigma_f$ or resistance against plastic deformation and particle distance distribution. The last-mentioned is
characterised by roughness height $h_r$ as well as molecular centre distance $a_{F=0}$. It corresponds to
an abscissa intersection $\sigma_{1,Z}$ of the constitutive consolidation function, Eq. (28) and Fig. 7.

**COHESIVE POWDER FLOW CRITERIA**

Using the elastic-plastic particle contact constitutive model Eq. (14) the failure conditions of
particle contacts are formulated, Tomas (2001a). It should be noted that the stressing pre-history
of a cohesive powder flow is stationary (steady-state) and delivers significantly a cohesive sta-
tionary yield locus in the $\tau$-$\sigma$-diagram of Fig. 5, Tomas (2000):

$$
\tau = \tan \varphi \cdot (\sigma + \sigma_0) \tag{17}
$$

This shear zone is characterised by a dynamic equilibrium of simultaneous contact shearing, un-
loading and failing, creating new contacts, loading, reloading, unloading and shearing again. The
stationary yield locus is the envelope of all Mohr-circles for steady-state flow (critical state line)
with a certain negative intersection of the abscissa

$$
\sigma_0 = \frac{1 - \varepsilon_0}{\varepsilon_0} \cdot \frac{F_{H,0}}{d^2} \tag{18}
$$

This isostatic tensile strength $\sigma_0$ of an unconsolidated powder without any particle contact de-
formation is obtained from the adhesion force $F_{H,0}$, Eq. (1), with the initial porosity of very loose
packing $\varepsilon_0 = 1 - \rho_{b,0} / \rho_s$, Eq. (34).
From the above formulation for cohesive steady-state flow Eq. (17), the stress-dependent effective angle of internal friction $\phi_e$ as introduced by Jenike (1964), i.e., slope of cohesionless effective yield locus, follows in accordance with experimental experience, Tomas (2000):

$$\sin \phi_e = \sin \phi_{st} \left( \frac{\sigma_1 + \sigma_0}{\sigma_1 - \sin \phi_u \cdot \sigma_0} \right)$$  \hspace{1cm} (19)

If the major principal stress $\sigma_1$ reaches the stationary uniaxial compressive strength $\sigma_{c, st}$, the effective angle of internal friction amounts to $\phi_e = 90^\circ$ and for $\sigma_1 \to \infty$ follows $\phi_e \to \phi_{st}$. For the combination of angle of internal friction $\phi_i$ for incipient contact failure (slope of yield locus) with the stationary angle of internal friction $\phi_{st}$ following relation is used, Molerus (1975) and again Tomas (2001a):

$$\tan \phi_{st} = (1 + \kappa) \cdot \tan \phi_i$$  \hspace{1cm} (20)

The softer the particle contacts, the larger are the differences between these friction angles and consequently, the more cohesive is the powder response.

The instantaneous yield locus describes the limit of incipient plastic powder deformation or yield. A linear yield locus, Fig. 5, is obtained from resolution of a general square function, see the details in Tomas (2001a), which is simply to use ($\sigma_{M, st}$, $\sigma_{R, st}$ centre and radius of Mohr circle for steady-state flow as parameter of powder pre-consolidation):

$$\tau = \tan \phi_i \cdot (\sigma + \sigma_Z) = \tan \phi_i \cdot \left( \sigma + \frac{\sigma_{R, st}}{\sin \phi_i} - \sigma_{M, st} \right)$$  \hspace{1cm} (21)

It is worth to note here that only the isostatic tensile strength $\sigma_Z$ for incipient yield depends directly on the consolidation pre-history and is given by:

$$\sigma_Z = \frac{\sigma_{R, st}}{\sin \phi_i} - \sigma_{M, st} = \left( \frac{\sin \phi_{st}}{\sin \phi_i} - 1 \right) \cdot \sigma_{M, st} + \frac{\sin \phi_{st}}{\sin \phi_i} \cdot \sigma_0$$  \hspace{1cm} (22)

The smaller a radius stress for pre-consolidation $\sigma_{VR} < \sigma_{R, st}$, the larger is the centre stress $\sigma_{VM} > \sigma_{M, st}$ right of largest Mohr circle for steady-state flow in Fig. 5, and the smaller would be the powder tensile strength $\sigma_Z$. The so-called consolidation locus lies at the right hand side and
represents the envelope of all Mohr circles for consolidation stresses with plastic powder failure, Fig. 5, i.e. the radius $\sigma_{VR}$ and centre $\sigma_{VM}$ stresses. Provided that the particle contact failure is equivalent to that between incipient powder flow and consolidation, one can write for a linear consolidation locus with negative slope $-\sin\phi_i$ which is symmetrically with the linear yield locus, Eq. (26):

$$\sigma_{VR} = \sin\phi_i \cdot (-\sigma_{VM} + \sigma_{iso})$$  \hspace{1cm} (23)

Due to this symmetry between yield and consolidation locus, one can directly estimate the isostatic powder compression $\sigma_1 = \sigma_2 = \sigma_{VM} = \sigma_{iso}$ from Fig. 5 for the radius stress $\sigma_{VR} = 0$:

$$\sigma_{iso} = 2 \cdot \sigma_{M,\text{st}} + \sigma_Z = \frac{\sigma_{R,\text{st}}}{\sin\phi_i} + \sigma_{M,\text{st}} = \left(\frac{\sin\phi_{\text{st}}}{\sin\phi_i} + 1\right) \cdot \sigma_{M,\text{st}} + \frac{\sin\phi_{\text{st}}}{\sin\phi_i} \cdot \sigma_0$$ \hspace{1cm} (24)

Generally, when we use these radius $\sigma_R$ and centre stresses $\sigma_M$, the essential flow parameters are compiled as one set of linear constitutive equations, i.e. for instantaneous consolidation, the consolidation locus (CL),

$$\sigma_R = \sin\phi_i \cdot (-\sigma_M + \sigma_{M,\text{st}}) + \sigma_{R,\text{st}}$$ \hspace{1cm} (25)

for incipient yield, the yield locus (YL),

$$\sigma_R = \sin\phi_i \cdot (\sigma_M - \sigma_{M,\text{st}}) + \sigma_{R,\text{st}}$$ \hspace{1cm} (26)

and for steady-state flow, the stationary yield locus (SYL):

$$\sigma_{R,\text{st}} = \sin\phi_{\text{st}} \cdot (\sigma_{M,\text{st}} + \sigma_0)$$ \hspace{1cm} (27)

These yield functions are completely described only with three material parameters plus the characteristic pre-consolidation stress $\sigma_{M,\text{st}}$ or average pressure influence, Tomas (2001a):

1. $\phi_i$ – incipient particle friction of failing contacts, i.e. Coulomb friction;
2. $\phi_{\text{st}}$ – steady-state particle friction of failing contacts, increasing adhesion by means of flattening of contact expressed by the contact consolidation coefficient $\kappa$, or by friction angles $(\sin\phi_{\text{st}} - \sin\phi_i)$ as shown in the next Eqs. (28) and (29). The softer the particle contacts, the larger are the difference between these friction angles the more cohesive is the powder;
(3) \( \sigma_0 \) – extrapolated isostatic tensile strength of unconsolidated particle contacts without any contact deformation, equals a characteristic cohesion force in an unconsolidated powder;

(4) \( \sigma_{M,\text{st}} \) – previous consolidation influence of an additional normal force at particle contacts, characteristic centre stress of Mohr circle of pre-consolidation state directly related to powder bulk density. This average pressure influences the increasing isostatic tensile strength of yield loci via the cohesive steady-state flow as the stress history of the powder.

All these physically based flow parameters are necessary to derive the uniaxial compressive strength \( \sigma_c \) which is simply found from the linear yield locus, Eq. (26) and Fig. 5, for \( \sigma_c = 2\sigma_R \) (\( \sigma_2 = 0 \) and \( \sigma_R = \sigma_M \)) as a linear function of the major principal stress \( \sigma_1 \), Fig. 7, Tomas (2000):

\[
\sigma_c = \frac{2 \cdot \left( \sin \varphi_{st} - \sin \varphi_i \right)}{(1 + \sin \varphi_{st}) \cdot (1 - \sin \varphi_i)} \cdot \sigma_1 + \frac{2 \cdot \sin \varphi_{st} \cdot \left( 1 + \sin \varphi_i \right)}{(1 + \sin \varphi_{st}) \cdot (1 - \sin \varphi_i)} \cdot \sigma_0 \quad (28)
\]

Equivalent to this linear function of the major principal stress \( \sigma_1 \) and using again Eq. (26), the absolute value of the uniaxial tensile strength \( \sigma_{Z,1} \) is also found for \( \sigma_{Z,1} = 2\sigma_R \) (\( \sigma_1 = 0 \) and \( \sigma_R = -\sigma_M \)), Tomas (2003):

\[
\sigma_{Z,1} = \frac{2 \cdot \left( \sin \varphi_{st} - \sin \varphi_i \right)}{(1 + \sin \varphi_{st}) \cdot (1 + \sin \varphi_i)} \cdot \sigma_1 + \frac{2 \cdot \sin \varphi_{st} \cdot \left( 1 + \sin \varphi_i \right)}{1 + \sin \varphi_{st}} \cdot \sigma_0 \quad (29)
\]

Both flow parameters \( \sigma_c \) and \( \sigma_{Z,1} \) depend on the pre-consolidation level of the shear zone which is expressed here by the applied consolidation stress for steady-state flow \( \sigma_1 \).

A considerable time consolidation under this major principal stress \( \sigma_1 \) after one day storage at rest is also shown in Fig. 7. Equivalent linear functions are also used to describe these time consolidation effects, Tomas (2001a).

**POWDER FLOWABILITY AND COMPRESSIBILITY**

In order to assess the flow behaviour of a powder, Eq. (28) shows that the flow function due to Jenike (1964) \( f^c_\theta = \sigma_1 / \sigma_c \) is not constant and depends on the pre-consolidation level \( \sigma_1 \). Approximately, one can write for a small intercept with the ordinate \( \sigma_{c,0} \), Fig. 7, the stationary angle of internal friction is equivalent to the effective angle \( \varphi_{st} \approx \varphi_e \) and Jenike’s (1964) formula is obtained:
Thus, the semi-empirical classification by means of the flow function introduced by Jenike (1964) is adopted here with considerations for certain particle behaviour, Table 1.

Obviously, the flow behaviour is mainly influenced by the difference between the friction angles, Eq. (30), as a measure for the adhesion force slope $\kappa$ in the general linear particle contact constitutive model, Eq. (14). Thus one can directly correlate $\kappa$ with flow function $f_{fc}$, Tomas (2001a):

$$
\kappa = \frac{1 + (2 \cdot f_{fc} - 1) \cdot \sin \phi_i}{\tan \phi_i \cdot (2 \cdot f_{fc} - 1 + \sin \phi_i)} \cdot \left(1 - \frac{1}{1 - \left(\frac{1 + (2 \cdot f_{fc} - 1) \cdot \sin \phi_i}{2 \cdot f_{fc} - 1 + \sin \phi_i}\right)^2 - 1}\right)^{-1}
$$

A characteristic value $\kappa = 0.77$ for $\phi_i = 30^\circ$ of a very cohesive powder is included in the adhesion force diagram, Fig. 4, and shows directly the correlation between strength and force increasing with pre-consolidation, Table 1. Due to the consolidation function, a small slope designates a free flowing particulate solid with very low adhesion level because of stiff particle contacts but a large slope implies a very cohesive powder flow behaviour because of soft particle contacts, Fig. 7.

Obviously, the finer the particles the “softer” are the contacts and the more cohesive is the powder, Tomas (1991, 2000). Köhler (1990) has experimentally confirmed this thesis for alumina powders ($\alpha$-$\text{Al}_2\text{O}_3$) down to the sub-micron range ($\sigma_{c,0} \approx \text{const.} = 2 \text{kPa}, d_{50} \text{median particle size in } \mu\text{m}$):

$$
f_{fc} \approx 2.2 \cdot d_{50}^{0.62}
$$

A survey of uniaxial compression equations was given by Kawakita (1970). Thus in terms of a moderate cohesive powder compression, to draw an analogy to the adiabatic gas law $p \cdot V^{*e} = \text{const.}$, a differential equation for isentropic compression of a powder $dS = 0$, i.e. remaining stochastic homogeneous (random) packing without a regular order in the continuum, is derived, beginning with:
The total pressure including particle interaction $p = \sigma_{M,\text{st}} + \sigma_0$ should be equivalent to a pressure term with molecular interaction $(p + a_{\text{vdW}}/V_m^2)(V_m - b) = R \cdot T$ in Van der Waals equation of state which is valid near gas condensation point. A “condensed” loose powder packing is obtained $\rho_b = \rho_{b,0}$, if only particles are interacting without an external consolidation stress $\sigma_{M,\text{st}} = 0$, e.g. particle weight compensation by a fluid drag, and Eq. (33) is solved:

$$\frac{p}{\rho_{b,0}} = \left(\frac{\sigma_0 + \sigma_{M,\text{st}}}{\sigma_0}\right)^n$$

(34)

Therefore, this physically based compressibility index $n \equiv 1/\kappa_{ad}$ lies between $n = 0$, i.e. incompressible stiff bulk material and $n = 1$, i.e. ideal gas compressibility. Considering the predominant plastic particle contact deformation in the stochastic homogeneous packing of a cohesive powder, following values of compressibility index are recommended in Table 2.

Generally, the influence of micro-properties as particle contact stiffness on the macro-behaviour as powder flow properties, i.e. cohesion, flowability and compressibility, can be directly shown in Fig. 8.

**CONCLUSIONS**

A complete set of physically based equations for steady-state flow, incipient powder consolidation and yielding, compressibility and flowability has been shown. Using this, the yield surfaces due to theory of plasticity may be described with very simple linear expressions:

$$\Phi_{\text{YL,SYL,CL}} = 0 = \begin{cases} 
\sigma_R - \sin \varphi_i \cdot (\sigma_M - \sigma_{M,\text{st}}) - \sigma_{R,\text{st}} & \text{yield locus (YL)} \\
\sigma_R - \sin \varphi_s \cdot (\sigma_{M,\text{st}} + \sigma_0) & \text{stationary yield locus (SYL)} \\
\sigma_R - \sin \varphi_i \cdot (\sigma_M + \sigma_{M,\text{st}}) - \sigma_{R,\text{st}} & \text{consolidation locus (CL)}
\end{cases}$$

(35)

The consolidation and yield loci and the stationary yield locus are completely described only with three material parameters, i.e., angle of internal friction $\varphi_i$, stationary angle of internal friction $\varphi_s$, isostatic tensile strength of an unconsolidated powder $\sigma_0$ plus the characteristic pre-consolidation (average pressure) influence $\sigma_{M,\text{st}}$. The compressibility index $n$ as an additional constitutive bulk powder parameter was introduced and the classification $0 \leq n < 1$
tutive bulk powder parameter was introduced and the classification $0 \leq n < 1$ recommended. A direct correlation between flow function $f \kappa_c$ and elastic-plastic contact consolidation coefficient $\kappa$ was derived.

This approach has been used to evaluate the powder flow properties concerning various particle size distributions (nanoparticles to granules), moisture contents (dry, moist and wet) and material properties (minerals, chemicals, pigments, waste, plastics, food etc.), which have been tested and evaluated for more than the last 20 years, Tomas (1991). Thus, these models are directly applied to evaluate the test data of a new oscillating shear cell, Kollmann (2001, 2002), Haack (2003), and a press-shear-cell in the high-level pressure range from 50 to 2000 kPa for liquid saturated, compressible filter cakes, Reichmann (2001, 2001a) and Mladenchev (2003), and for dry powders, Grossmann (2003). Additionally, the force – displacement behaviour during stressing and the breakage probability are useful constitutive functions to describe the mechanics of agglomerates to assess the physical product quality, Antoniuk (2003). These contact models are also needed to simulate the shear dynamics of cohesive powders using the discrete element method (DEM) and to calibrate these simulations by shear cell measurements, Tykhoniuk (2003).

The influence of particle surface properties, e.g. as contact stiffness, on the powder flow properties can be directly interpreted, Fig. 8, and practically used to design particulate products in process industries.

**Symbols**

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<td>a</td>
<td>separation, nm</td>
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<tr>
<td>A</td>
<td>area, particle contact area, m²</td>
</tr>
<tr>
<td>$C_H$</td>
<td>Hamaker constant, J</td>
</tr>
<tr>
<td>d</td>
<td>particle size, µm</td>
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<tr>
<td>E</td>
<td>modulus of elasticity, kN/mm²</td>
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<tr>
<td>F</td>
<td>force, N</td>
</tr>
<tr>
<td>p</td>
<td>pressure, kPa</td>
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<tr>
<td>$p_f$</td>
<td>plastic yield strength of particle contact, MPa</td>
</tr>
<tr>
<td>r</td>
<td>radius, nm</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>Porosity</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>elastic-plastic contact consolidation coefficient</td>
</tr>
<tr>
<td>$\kappa_A$</td>
<td>elastic-plastic contact area coefficient</td>
</tr>
<tr>
<td>$\kappa_p$</td>
<td>plastic contact repulsion coefficient</td>
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**Indices**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>b</td>
<td>bulk</td>
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<tr>
<td>c</td>
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<tr>
<td>K</td>
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<td>effective</td>
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<td>M</td>
<td>centre of Mohr circle</td>
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<td>N</td>
<td>normal</td>
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<tr>
<td>pl</td>
<td>plastic</td>
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<tr>
<td>R</td>
<td>radius of Mohr circle</td>
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<td>s</td>
<td>solid</td>
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</tbody>
</table>
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| Table 1: Flowability assessment and elastic-plastic contact consolidation coefficient $\kappa$($\varphi_i = 30^\circ$) |
|---|---|---|---|---|
| flow function $f_{f_c}$ | $\kappa$-values | $\varphi_{st}$ in deg | evaluation | Examples |
| 100 - 10 | 0.01 - 0.107 | 30.3 - 33 | free flowing | dry fine sand |
| 4 - 10 | 0.107 - 0.3 | 33 - 37 | easy flowing | moist fine sand |
| 2 - 4 | 0.3 - 0.77 | 37 - 46 | cohesive | dry powder |
| 1 - 2 | 0.77 - $\infty$ | 46 - 90 | very cohesive | moist powder |
| $< 1$ | $\infty$ | - | non flowing | moist powder |

| Table 2: Compressibility index of powders, semi-empirical estimation for $\sigma_1 = 1 – 100$ kPa |
|---|---|---|---|
| index $n$ | evaluation | examples | flowability |
| 0 – 0.01 | incompressible | gravel | free flowing |
| 0.01 – 0.05 | low compressibility | fine sand | |
| 0.05 - 0.1 | compressible | dry powder | cohesive |
| 0.1 - 1 | very compressible | moist powder | very cohesive |
a) nonlinear elastic, adhesion

b) linear plastic, adhesion, dissipative

c) nonlinear viscoelastic

d) nonlinear plastic

e) nonlinear elastic, dissipative

f) linear viscoplastic

g) linear plastic, nonlinear elastic, dissipative, adhesion

h) linear plastic, nonlinear elastic, adhesion, dissipative, viscoelastic, viscoplastic

Fig. 1: Constitutive models of contact deformation of smooth spherical particles in normal direction without (only compression +) and with adhesion (tension -). The basic models for elastic behavior were derived by Hertz (1882), for viscoelasticity by Yang (1966), for constant adhesion
by Johnson et al. (1985) and for plastic behaviour by Thornton and Ning (1998) and Walton and Braun (1986) and for plasticity with variation in adhesion by Molerus (1975) and Schubert et al. (1976). This has been expanded stepwise to include nonlinear plastic contact hardening and softening. Energy dissipation was considered by Sadd et al. (1993) and time dependent viscoplasticity by Rumpf et al. (1976). Considering all these theories, one obtains a general contact model for time and rate dependent viscoelastic, elastic-plastic, viscoplastic, adhesion and dissipative behaviours, Tomas (2000, 2001).
Fig. 2: Particle contact approach, elastic, elastic-plastic deformation and detachment. After loading with an external force $F_N$ the spherical contact is elastically compacted to a plate-plate-contact and shows the Hertz (1882) elliptic pressure distribution b). With increasing normal load this contact starts at the yield point $p_{\text{max}} = p_f$ with plastic yielding c). The micro-yield surface is reached and this maximum pressure has not been exceeded. A hindered plastic field is formed at the contact with a circular constant pressure $p_{\text{max}}$ and an annular elastic pressure distribution dependent on radius $r_{K,\text{el}}$, full lines in c).
Fig. 3: Recalculated characteristic contact deformation of very cohesive titania particles, surface diameter $d_S = 200$ nm, surface moisture $X_W = 0.4\%$. The origin of this diagram $h_K = 0$ is equivalent to the characteristic adhesion separation for direct contact $a_{F=0}$. After loading $0 - Y$ the contact is elastically compacted with an approximated circular contact area, Fig. 2b) and starts at the yield point $Y$ at $p_{\text{max}} = p_r$ with plastic yielding, Fig. 2c). Next, the combined elastic-plastic yield boundary of the plate-plate contact is achieved, Eq. (3). This displacement is expressed by annular elastic $A_{el}$ (thickness $r_{K,el}$) and circular plastic $A_{pl}$ (radius $r_{K,pl}$) contact area, Fig. 2c).
Fig. 4. Recalculated particle contact forces of titania according Eq. (14) using data of Fig. 6, surface diameter $d_s = 200$ nm, surface moisture $X_W = 0.4$ %. The points characterise the pressure levels of YL 1 to YL 4 according to Fig. 7. A characteristic line for $\kappa = 0.77$ of a very cohesive powder is included and shows directly the correlation between strength and force enhancement with pre-consolidation, Eq. (31).
Fig. 5. Yield characteristics of a cohesive powder in the $\tau$-$\sigma$-diagram. In general, the steady-state flow of a cohesive powder is cohesive. Hence, the total normal stress consists of an external contribution $\sigma$, e.g. by weight of powder layers, plus (by absolute value) an internal contribution by pre-consolidation dependent adhesion (tensile stress $\sigma_z$).
Fig. 6: Yield loci (YL) and stationary yield locus (SYL) of titania powder, straight line regression fit $\geq 0.97$, $d_S = 200$ nm, solid density $\rho_s = 3870$ kg/m$^3$, shear rate $v_S = 2$ mm/min, surface moisture $X_W = 0.4\%$; angles of internal friction $\phi_i = 25 - 37^\circ$, stationary angle of internal friction $\phi_i = 54^\circ$, isostatic tensile strength $\sigma_0 = -0.33$ kPa
Fig. 7. Constitutive consolidation function of titania, straight line regression fit = 0.99, $d_s = 200$ nm, surface moisture $X_W = 0.4$ % accurately analysed by Karl Fischer titration.
Fig. 8: Characteristic constitutive functions of stiff and compliant particle contact behaviours, free flowing and cohesive powder behaviours, and finally, stiff incompressible and soft compressible powders, Tomas (2002). Increasing contact compliance determine decreasing slope of the elastic-plastic yield boundary (limit) and increasing inclination of the adhesion boundary or limit. As the result, the slope of the normal force-adhesion force function increases. Next, the difference between the stationary angle and angle of internal friction of the powder becomes larger. Consequently, the slope of the powder consolidation function increases and the powder is more compressible.