Mechanics of Particle Adhesion

Jürgen Tomas
Mechanical Process Engineering, Department of Process Engineering and Systems Engineering, Otto-von-Guericke-University, Universitätsplatz 2, D – 39106 Magdeburg, Germany

Abstract
The fundamentals of particle-particle adhesion for ultrafine particles (d < 10 µm) are presented using continuum mechanics approaches. The models for elastic (Hertz, Huber), elastic-adhesive (Derjaguin, Johnson, Maugis, Greenwood), viscoelastic (Yang), plastic-adhesive (Krupp, Molerus, Johnson, Maugis & Pollock) contact displacement response of a single, normal loaded, isotropic, smooth contact of two spheres are discussed. The force-displacement behaviours of elastic-plastic (Schubert, Thornton), elastic-dissipative (Sadd), plastic-dissipative (Walton), plastic-hardening (Vu-Quoc, Castellanos) and viscoplastic-adhesive (Rumpf) contacts are also shown. Hook’s linear elastic tangential force-displacement relation, non-linear contact loading paths up to Coulomb friction as its limit (Fromm, Föppl, Sonntag, Mindlin), the effect of adhesion (Derjaguin, Thornton) and different non-linear paths for load, unload, reload, reverse shear load, unload and load (Mindlin & Deresiewicz, Walton & Braun, Thornton) are compiled in a comprehensive literature review.

Based on these theories, a general approach for the time and deformation rate dependent and combined viscoelastic, plastic, viscoplastic, adhesion and dissipative behaviours of a contact between spherical particles is derived and explained. Using the new model “stiff particles with soft contacts”, the influence of elastic-plastic repulsion in a “representative particle contact” (RPC) is shown by a contact force equilibrium. The attractive particle adhesion contribution is described by surface forces, i.e. van der Waals forces. A sphere-sphere adhesion model without any contact deformation is combined with a plate-plate adhesion model for the nanocontact flattening. Using this model, various contact deformation paths for loading, unloading, reloading and contact detachment are comfortably to discuss. Thus, the varying adhesion forces between particles depend directly on this “frozen” irreversible deformation, the so-called contact history. The contribution of this history or load dependent adhesion on the tangential force in an elastic-plastic frictional contact is derived. With this increasing flattening, normal and tangential contact stiffness, rolling and twisting resistance, energy absorption and friction work increase. For colliding particles with adhesion, the correlation between particle impact velocity and contact deformation response is obtained using energy balance. Thus, the constitutive model approach is generally applicable for solid micro- or nanocontacts but has been shown here for ultrafine limestone particles (median particle size d_{50,3} = 1.2 µm) as typical example of a cohesive powder.

Keywords: Contact mechanics, constitutive models, adhesion force, ultrafine particles, powder

1 Extended master version of [181] and full version of review paper of CHISA event, Prague 2004
2 Phone: ++49 391 67 18783, Fax: ++49 391 67 11160, e-mail: juergen.tomas@vst.uni-magdeburg.de
MECHANICS OF PARTICLE ADHESION

1. INTRODUCTION

2. PARTICLE ADHESION EFFECTS
   2.1 PARTICLE BOND EFFECTS
   2.2 COMPARISON BETWEEN DIFFERENT ADHESION FORCES

3. PARTICLE CONTACT CONSTITUTIVE MODELS
   3.1 THE STATE OF ARTS
   3.2 ELASTIC, PLASTIC AND VISCOPLASTIC CONTACT DEFORMATIONS
   3.2.1 Elastic normal force-displacement function
   3.2.2 Elastic-frictional tangential force-displacement function
   3.2.3 Rolling resistance and elastic-frictional twisting of an elastic particle contact
   3.2.4 Elastic displacement of an adhesive contact
   3.2.5 Perfect plastic and viscoplastic contact displacement and viscous damping
   3.3 LOAD DEPENDENT ADHESION FORCE
   3.3.1 Increase of adhesion force by contact consolidation
   3.3.2 Linear increase of adhesion force by normal force
   3.3.3 Non-linear increase of adhesion force by normal force
   3.4 VARIATION IN ADHESION DUE TO INELASTIC CONTACT FLATTENING
   3.4.1 Elastic-plastic force-displacement model for loading
   3.4.2 Unloading and reloading hysteresis and contact detachment
   3.4.3 Viscoplastic contact behaviour and time dependent consolidation
   3.5 MEASUREMENT OF ADHESION FORCES
   3.5.1 Adhesion effects between particles and surfaces
   3.5.2 Principles of adhesion force testing methods
   3.6 ENERGY ABSORPTION IN A CONTACT WITH ADHESION AND PARTICLE IMPACT
   3.6.1 Dissipative behaviour of contact unload/reload
   3.6.2 Elastic particle impact and maximum impact force
   3.6.3 Coefficient of restitution at particle impact
   3.6.4 Particle impact and adhesion, contact displacement and impact force response
   3.7 REPRESENTATIVE, LOAD DEPENDENT ADHESION FORCE
   3.7.1 Linear adhesion force – normal force function
   3.7.2 Adhesion force of a stiff contact with surface asperity
   3.7.3 Non-linear adhesion force – normal force function
   3.7.4 Elastic-plastic, frictional tangential force of load dependent adhesion contact
   3.7.5 Elastic-plastic, frictional rolling resistance of load dependent adhesion contact
   3.7.6 Elastic-plastic, frictional twisting of load dependent adhesion contact

4. CONTACT FAILURE BY SHEAR AND POWDER YIELD

5. CONCLUSIONS
1. Introduction

In particle processing and product handling of fine (d < 100 µm), ultrafine (d < 10 µm) and nanosized particles (d < 0.1 µm), the well-known flow problems of cohesive powders in process apparatuses or storage and transportation containers include bridging, channelling, widely spread residence time distribution associated with time consolidation or caking effects, chemical conversions and deterioration of bioparticles. Avalanching effects and oscillating mass flow rates in conveyors lead to feeding and dosing problems. Finally, insufficient apparatus and system reliability of powder processing plants are also related to these flow problems.

The rapid increasing production of cohesive to very cohesive ultrafine to nanosized powders [1], e.g. pigment particles, toner, pharmaceuticals, micro-carriers in biotechnology or medicine, auxiliary materials in catalysis, chromatography or silicon wafer polishing, make these problems much more serious. Taking into account this list of adhesion effects, technical problems and hazards in processing units, it is essential to deal with the fundamentals of particle adhesion, powder consolidation, compaction and flow, i.e. to develop a reasonable combination of particle and continuum mechanics, Fig. 1.

In terms of powder mechanics (see level I), the well-known failure hypotheses of Tresca, Coulomb-Mohr and Drucker-Prager (in [2 - 5]), the yield locus concepts of Jenike [6, 7] and Schwedes [8, 9], the Warren-Spring equations [10 - 14] and the approach by Tüzün [16, 17] were supplemented by Molerus [18 - 21] to describe the cohesive, steady-state powder flow criterion. Forces acting on particles under stress in a regular assembly and its dilatancy were considered by Rowe [22] and Horne [23]. Savage and Sayed [24], Johnson and Jackson [25], Nedderman [26, 27], Satake and Jenkins [28] have modelled the rapid and collisional flow of non-adhering particles. Moreau [29], Bagi [30, 31], Kruyt [32, 33], Goddard [34], Satake [35], Kaneko et al. [36], Goldhirsch [37], Radjai [38] and others described the micromechanics of granular media. Tardos [39] discussed the frictional flow for compressible powders without any cohesion from the fluid mechanics point of view.

Moreover, modelling and simulation of particle dynamics of free flowing granular media is increasingly used, see e.g. Deresiewicz [40], Cundall [41], Campbell [42], Walton [43, 44], Duran [45], Herrmann [46, 47], Thornton [48], Luding [49, 50] and Wolf [51 - 53].

The fundamentals of contact mechanics and impact mechanics are compiled and described by Johnson [54] and Stronge [55]. Multibody dynamics, their contacts and finite element modelling are shown by Eberhard [56].
The fundamentals of molecular attraction potentials and the mechanics of adhesion are treated for example by Krupp [57], Johnson [54], Israelachvili [58], Maugis [59] and Kendall [60].

Fig. 1: Hierarchy of the dynamics of random packing in particle beds, clusters or agglomerates (fixed spatial coordinates $x_1, x_2, x_3$, $i,j = 1, 2, 3$; normal stresses $\sigma_{ii}$ and shear stresses $\sigma_{ij}$, compression $v_{ii}$ and shear rates $v_{ij}$, normal strains $\varepsilon_{ii}$, distortions $\varepsilon_{ij}$, stress increments $\Delta \sigma_{ij}$ and $d\sigma_{ij}$).

I. Level of Continuum Mechanics: 3-dimensional continuum models described by tensor equations ($2^{nd}$ order vectors) of representative volume elements (RVE). This includes balances for mass (continuity), forces, momentum (impulse), moment (torque) and energy for 3 translational and 3 rotational degrees of freedom.

II. Level of Micromechanics: Macro – microtransition to a representative, geometrically equivalent, 2-dimensional plane element with a finite number of discrete sub-elements, i.e. particles. All the particle interactions inside of a random packing structure are described by contact forces ($1^{st}$ order vectors) in normal and tangential direction with fundamental laws for elastic force-displacement relations, inelastic deformations (plastic displacements), solid friction and viscous damping – see the typical symbols of spring, Coulomb-friction and dashpot elements in the circular detail.

III. Level of Molecular Dynamics: Dominant molecular interactions at particle contact surfaces. All the mentioned mechanical properties are influenced by surface properties, e.g. adsorbed layers of condensed matter (adsorbed moisture, electrostatic bilayers or surfactants). Generally, fundamental material properties as elasticity, bonding strength, viscosity (rate dependent stiffness), electromagnetic, thermal and mechanical wave propagation characteristics or phase conversion enthalpies can be physically explained by these molecular interaction energies and potential forces and simulated by molecular dynamics.
In particle technology, adhesion effects are related to undesired powder blocking at conveyor transfer chutes [61], in pneumatic pipe bends [62], to fluidization problems in handling [63], to desired particle cake formation on filter media [64, 65] or to pigment adhesion. The caking of food [66, 67], toner handling [68], wear effects of adhering solid surfaces [69, 70], fouling at heat exchangers [71] or in membrane filters [72], thermal conductivity in particle beds [73, 74], fine particle deposition in lungs, formulation of particulate products, particle coating, powder tabletting and briquetting, and the granulate strength [75 - 86] or surface cleaning of silicon wafers [87 - 95], etc., are also connected with adhesion - desired or undesired. Adhesion and friction is also an serious problem dealt in tribology [15, 16].

First, the essential particle bond effects are introduced and compared in chapter 2. In the main chapter 3, after a comprehensive literature review 3.1, the elastic, plastic and viscoplastic force – displacement models are explained in section 3.2. Then the problem of load dependent adhesion force is treated by reviewing the literature, section 3.3. Because of essential problems related to ultrafine particles the elastic-plastic yield and adhesion limits of contact flattening are derived as quasi-static problems in section 3.4. Measurements, energy balances and impact problems of rapid moving particles are treated in sections 3.5 and 3.6. The consequences for the representative load dependent adhesion force with respect to contact sliding, particle rolling and torsion, Fig. 2, can be read in section 3.7. How to apply these contact laws to model the consolidation and frictional shear flow of ultrafine cohesive powders, i.e. for applications in powder processing, storage and handling, is seen in chapter 4.

![Fig. 2: Particle contact forces, moments and degrees of freedom: one normal load or central impact in direction of principal axis and one torsional moment around this axis, two tangential forces and two rolling moments within the plane of flattened contact and their respective constitutive force – displacement and moment – angle relations.](Mechanics_Particle_Adhesion_full.doc)
2. Particle adhesion effects

2.1 Particle bond effects

Particle adhesion can be explained by following bond effects, Fig. 3, [75 - 81, 96 - 100]:

- **Surface and field forces at direct contact**
  - Van der Waals forces
  - Electrostatic forces
    - *Conductor*
    - *Non-conductor*
  - Magnetic force

- **Material bridge between contacts**
  - Organic macromolecules (floculants)
  - Liquid bridge bonds
    - *Low viscosity*
    - *High viscosity*
  - Solid bridge bonds
    - Recrystallisation of liquid bridges
    - Contact fusion by sintering
    - Chemical solid-solid reaction

- **Interlocking by hook-like bonds**

Fig. 3: Particle adhesion and microprocesses of particle bond effects in contact:

- Surface and field forces at direct contact:
  - Van der Waals forces,
  - Electrostatic forces,
    - electric conductor,
    - electric non-conductor,
  - Magnetic force;
- Material bridges between particle surfaces:
  - Hydrogen bonds of adsorbed surface layers of condensed water,
  - Organic macromolecules as floculants in suspensions;
  - Liquid bridges of
    - Low viscous wetting liquids by capillary pressure and surface tension,
    - High viscous bond agents (resins);
  - Solid bridges by
Recrystallisation of liquid bridges which contains solvents (salt),
- Solidification of swelled ultrafine sol particles (starch, clay),
- Freezing of liquid bridge bonds,
- Chemical reactions with adsorbed surface layers (cement hydration by water),
- Solidification of high viscous bond agents (asphalt),
- Contact fusion by sintering (aggregates of nanoparticles),
- Chemical bonds by solid-solid reactions (glass batch);
- Interlocking by macromolecular and particle shape effects;
  - Interlocking of chain branches at macromolecules (proteins),
  - Interlocking of contacts by overlaps of surface roughness,
  - Interlocking by hook-like bonds.

These bond effects are directly related to the micromechanical level II and III explained before.

2.2 Comparison between different adhesion forces

The strength of different adhesion effects is compared in Fig. 4. Disregarding the solid bridge bonds [96 - 98, 159, 328], the liquid bridge is the dominating adhesion force [77], as long as the liquid bridge is formed, see e.g. Tomas [96], Schubert [101], Adams [102], Ennis [103] or Seville [104].

Fig. 4: Adhesion forces between stiff particle and smooth surface according to Rumpf [77], calculated with \( a_0 = 0.4 \) nm molecular centre separation (at interaction force equilibrium), \( \alpha = 20^\circ \) bridge angle, \( \theta = 0^\circ \) wetting angle, \( \sigma_{lg} = 72 \) mJ/m\(^2\) surface tension of water, \( C_H = 1910^{20} \) J Hamaker constant acc. to Lifshitz, \( q_{max} = 16 \times 10^{-18} \) As/\( \mu m^2 \) surface charge density, \( U = 0.5 \) V contact potential, \( C_{H,ds} = \left( \sqrt{C_{H,as}} - \sqrt{C_{H,ls}} \right) \) Hamaker constant for particle-water-particle interaction. Instead of the smooth plate one can also consider a coarse smooth particle with a large radius of surface curvature.
Without the liquid bridge, the van der Waals force of a dry contact dominates at fine particles. This van der Waals force can be considerably decreased in wet environment because of the reduction of Hamaker constant by the interstitial water. This effect is widely used in washing processes. For charged particles such as toner particles the Coulomb force becomes important. The surface-charge density is assumed to be \( q_{\text{max}} = 16 \times 10^{-18} \text{ As/\mu m}^2 \) which is the maximum value determined by the electric field limit for discharging. Sometimes the Coulomb force may be larger than the van der Waals force because the maximum surface-charge density is determined by the voltage limit rather than the field limit [105].

In the above discussion, the effects of the atmospheric conditions are not taken into consideration. Because of humidity of the ambient air water is adsorbed at particle surfaces. These surface layers of condensed water increase remarkably the effective contact zone. By molecular bonds between these condensed liquid molecules (known as surface tension), these surface layers may form additionally small liquid bridges which results in the alternation of the adhesion force [75, 106]. The humidity also changes the adsorbed water layer thickness, see e.g. [106 - 97], and it also affects the adhesion force. Thus, the adhesion is influenced by mobile adsorption layers due to molecular rearrangement and additional hydrogen bonds [100].

As long as the liquid bridge is stable, the critical separation for rupture is about \( a_{\text{crit}} \sim \frac{1}{\sqrt{h}} \) [107], the separation influences hardly the bond force. Van der Waals forces and Coulomb forces (for a conductor) have equivalent long-range force-separation (distance) curves \( F_{\text{H0}}(a) \), Fig. 4 middle panel. The Coulomb forces of a non-conductor do not depend on separation. This is widely used to precipitate dust particles in an electric field.

The effect of van der Waals forces strongly depends on the roughness of a surface, Fig. 4 panel b). Minimum of van der Waals force is seen versus roughness height \( h_r \) for different particle sizes \( d \). The influence of roughness for liquid bridge bonds and adsorption layers (humidity) is comparatively small. The adhesion will also be affected by temperature [105]. It is worth to note here that the weight of fine particles (\( d < 100 \mu m \)) is very small compared to the adhesion forces which dominate particle interaction in the gravitational field.

The force-displacement behaviours of elastic, elastic-adhesive, plastic-adhesive, elastic-plastic, elastic-dissipative, plastic-dissipative and viscoplastic-adhesive contacts are shown. Based on these individual theories, a general approach for the time and deformation rate dependent and combined viscoelastic, plastic, viscoplastic, adhesion and dissipative behaviours of a contact between spherical particles is derived and explained. We focus here on the adhesion forces at particle contacts which are necessary to understand physically the consolidation and non-rapid flow of more or less dry, fine, ultrafine down to nanoscaled cohesive powders (particle size \( d < 100 \mu m \)).
3. Particle contact constitutive models

3.1 The state of arts

An appropriate introduction in contact mechanics one can find at Fischer-Cripps [5] with respect to the fundamentals of engineering mechanics. Moreover, for particle technology, powder processing and handling, in principle, there are four essential deformation effects in particle-surface contacts and their force-response (stress-strain) behaviours can be explained as follows, Table 1:

(1) **elastic** contact deformation (Hertz [108], Huber [109], Derjaguin [110], Bradley [112], Fromm [113], Cattaneo [114], Föppl [115], Sonntag [116], Mindlin [117], Deresiewicz [122], Lurje [123], Sperling [124], Krupp [57], Greenwood [125], Johnson [126], Dahneke [127], Maw [129], Tsai [130], Thornton [131, 132], Sadd [133], Tavares [134], Vu-Quoc [135] or Di Renzo [141]) which is reversible, independent of deformation rate and consolidation time effects and valid for all particulate solids;

(2) **plastic** contact deformation with adhesion (Derjaguin [110], Krupp [142], Greenwood [125], Schubert [143], Molerus [18, 19], Maugis [144], Walton [145], Fleck [146], Greenwood [147], Thornton [150, 151], Tomas [152 - 155], Vu-Quoc [136, 139], Mesarovic & Johnson [156], Luding [157, 158], Delenne [159], Martin [160] or Castellanos [161]) which is irreversible, deformation rate and consolidation time independent, e.g. mineral or metal powders;

(3) **viscoelastic** contact deformation (Pao [162], Lee [163], Hunter [164], Yang [165], Krupp [57], Rumpf [166], Stieß [167], Walton [44], Sadd [133], Scarlett [169], Leszczynski [170] or Brilliantov [172]) which is reversible and dependent on deformation rate and consolidation time, e.g. plastics, soft organic chemicals or bio-particles;

(4) **viscoplastic** contact deformation (Rumpf [166], Stieß [167], Kuhn [168], Bouvard [173], Storakers [174, 175], Parhami [176, 177], Tomas [178 - 181], Fleck [182], McMeeking [183], Tomas [184, 185] or Luding [186]) which is irreversible and dependent on deformation rate and consolidation time, e.g. nanoparticles fusion.

In terms of simulations of particle dynamics by discrete element method the commonly used linear spring-dashpot model [41, 191] was extended according to viscous damping by Pöschel et al. [193], for plasticity by Luding [157, 158] and for liquid bridge bonds by Thornton [107] and Gröger [196, 197].
<table>
<thead>
<tr>
<th>authors</th>
<th>assumptions</th>
<th>characteristic force-displacement or adhesion force models</th>
<th>contact area and characteristic diagrams</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hertz [108] 1882</td>
<td>elastic behaviour, normal force $F_N$</td>
<td>$\left( \frac{p_{el}}{p_{max}} \right)^2 = 1 - \left( \frac{r_k}{r_{K,el}} \right)^2$ with $p_{max} = \frac{3 \cdot F_N}{2 \cdot \pi \cdot r_{K,el}^2}$</td>
<td><img src="Mechanics_Particle_Adhesion_full.doc" alt="Pressure" /></td>
</tr>
<tr>
<td></td>
<td></td>
<td>$F_N = \frac{2}{3} \cdot E^* \cdot \sqrt{r_{1,2} \cdot h_K^3}$</td>
<td><img src="Mechanics_Particle_Adhesion_full.doc" alt="Force" /></td>
</tr>
<tr>
<td>Huber [109] 1904</td>
<td>elastic behaviour, principal stresses</td>
<td>$\frac{\sigma_y(r_k)}{p_{max}} = \frac{1 - 2 \cdot \nu \cdot r_{K,el}^2}{3} \cdot \frac{r_k^2}{r_k^2} \cdot \left[ 1 - \left( \frac{r_k^2}{r_{K,el}^2} \right)^{3/2} \right] + \left( \frac{2}{3} \cdot \frac{r_k^2}{r_{K,el}^2} \right)^{3/2}$ for $r_K \leq r_{K,el}$</td>
<td><img src="Mechanics_Particle_Adhesion_full.doc" alt="Pressure" /></td>
</tr>
<tr>
<td>Fromm [113] 1927</td>
<td>elastic rolling disks, no-slip and partial-slip</td>
<td>$F_T = \mu_i \cdot F_N \cdot \left[ 1 - \left( \frac{E^<em>}{4 \cdot \mu_i \cdot p_{max, disk} \cdot b_{K,el}} \right) \cdot \delta^2 \right]$ slide: $\Delta V_{\nu} = \Delta \omega \approx \delta$ with $p_{max, disk} = \frac{2 \cdot F_N}{\pi \cdot b_{K,el}^2}$ and $b_{K,el} = \frac{8 \cdot r_{1,2} \cdot F_N}{\pi \cdot E^</em>}$</td>
<td><img src="Mechanics_Particle_Adhesion_full.doc" alt="Rolling" /></td>
</tr>
<tr>
<td>Föppl [115] 1936/1947 and Sonntag [187] 1950</td>
<td>elastic rolling, no-slip, partial-slip, friction</td>
<td>$F_T = \mu_i \cdot F_N \cdot \left[ 1 - \left( \frac{2 \cdot E^* \cdot r_{K,el} \cdot \delta}{3 \cdot \mu_i \cdot F_N} \right)^{5/2} \right]$ sliding at rolling</td>
<td><img src="Mechanics_Particle_Adhesion_full.doc" alt="Rolling" /></td>
</tr>
<tr>
<td>Author(s)</td>
<td>Year(s)</td>
<td>Description</td>
<td></td>
</tr>
<tr>
<td>-------------------</td>
<td>---------</td>
<td>-----------------------------------------------------------------------------</td>
<td></td>
</tr>
<tr>
<td>Cattaneo [114]</td>
<td>1938 and Mindlin [117] 1949</td>
<td>elastic behaviour, tangential slip $F_T$, shear stress by sliding $\tau$ and torsion $\tau_{\text{to}}$, [ F_T = 4 \cdot G \cdot r_K \cdot \delta ] for elastic no-slip, [ \delta = \frac{3 \cdot \mu \cdot F_N}{8 \cdot G \cdot r_{K,el}} \left[ 1 - \left( 1 - \frac{F_T}{\mu \cdot F_N} \right)^{2/3} \right] ] partial-slip: $r_{K,slip} \leq r_K \leq r_{K,el}$, [ \delta = \frac{3 \cdot \mu \cdot F_N}{8 \cdot G \cdot r_{K,el}} \left[ 2 \left( 1 - \frac{F_{T, U} + F_T}{2 \cdot \mu \cdot F_N} \right)^{2/3} - \left( 1 - \frac{F_{T, U}}{\mu \cdot F_N} \right)^{2/3} - 1 \right] ]</td>
<td></td>
</tr>
<tr>
<td>Mindlin and Deresiewicz [118] 1953</td>
<td>elastic behaviour, tangential unload and reload hysteresis, [ \tau = \frac{3 \cdot \mu \cdot F_N}{2 \cdot \pi \cdot r_{K,el}^2} \left( \sqrt{1 - \frac{r_K^2}{r_{K,el}^2}} - \sqrt{1 - \frac{r_{K,slip}^2}{r_{K,el}^2}} \right) ] partial-slip: $r_{K,slip} \leq r_K \leq r_{K,el}$, [ \delta = \frac{3 \cdot \mu \cdot F_N}{8 \cdot G \cdot r_{K,el}} \left[ 2 \left( 1 - \frac{F_{T, U} + F_T}{2 \cdot \mu \cdot F_N} \right)^{2/3} - \left( 1 - \frac{F_{T, U}}{\mu \cdot F_N} \right)^{2/3} - 1 \right] ]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lubkin [119] 1951, Cattaneo [120] 1952 and Deresiewicz [122] 1954</td>
<td>Elastic-frictional twisting, unload, reload hystereses, [ r_{K,\text{counterslip}} = \frac{r_{K,el}}{\sqrt{3}} \cdot \sqrt{4 \left( 1 - \frac{3 \cdot (M_{to} - M_{to}^*)}{4 \cdot \mu \cdot F_N \cdot r_{K,el}} \right)^{1/2} - 1} ]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sonntag [116] 1949, Hamilton [188] 1966</td>
<td>elastic sliding spheres, [ \sigma_y = \frac{3 \cdot F_{N}}{2 \cdot \pi \cdot r_{K,el}^2} \left[ 2 \cdot v \cdot C_0 + \left( 1 - 2 \cdot v \right) \left( \frac{C_1}{r_K^2} - \frac{2 \cdot v^2 \cdot C_1}{r_K^4} - \frac{y^2 \cdot C_0}{r_K^2} \right) \right] ] for $r_K &lt; r_{K,el}$, [ \tau = \frac{3 \cdot F_{N}}{2 \cdot \pi \cdot r_{K,el}^2} \left[ x \cdot C_0 \cdot \frac{2 \cdot y \cdot C_1}{r_K^2} - \frac{2 \cdot y \cdot C_1}{r_K^2} \right] ]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Author/Era</td>
<td>Description</td>
<td>Equation</td>
<td></td>
</tr>
<tr>
<td>------------</td>
<td>-------------</td>
<td>----------</td>
<td></td>
</tr>
<tr>
<td>Yang [165] 1966</td>
<td>Viscoelastic relaxation, normal force</td>
<td>[ r_k^3 = \frac{3 \cdot r_{1,2} \cdot F_N}{2 \cdot E_\infty^* (t = 0)} \left[ \frac{E_0^<em>}{E_\infty^</em> (t \to \infty)} + \left( 1 - \frac{E_0^<em>}{E_\infty^</em>} \right) \exp(-t/t_{\text{relax}}) \right] ]</td>
<td></td>
</tr>
</tbody>
</table>
| Johnson, Kendall and Roberts [126] 1971 | Elastic behaviour, normal force, constant adhesion | \[ r_k^3 = \frac{3 \cdot r_{1,2} \cdot \left( F_N + F_{H,JKR} + \sqrt{2 \cdot F_{H,JKR} \cdot F_N + F_{H,JKR}^2} \right)}{2 \cdot E^*} \]
\[ F_{H,JKR} = 6 \cdot \pi \cdot r_{1,2} \cdot \sigma_{ss} \] |
| Dahneke [127] 1972 | Elastic behaviour, normal force, varying adhesion | \[ F_H = F_{H0} + A_K \cdot p_{vaw} = \frac{C_H \cdot r_{1,2}}{6 \cdot a_0^2} \left( 1 + \frac{2 \cdot h_k}{a_0} \right) \]
\[ F_{H,\text{max}} = \frac{C_H \cdot r_{1,2} \cdot \left( 1 + \frac{2 \cdot C_H^2 \cdot r_{1,2} \cdot h_k}{27 \cdot E^* \cdot a_0^2} \right)}{6 \cdot a_0^2} \] |
| Derjaguin [110] 1934, Muller and Toporov [128] 1975 | Elastic behaviour, normal force, constant adhesion \( F_{H0} \) | \[ F_{H0} = 4 \cdot \pi \cdot r_{1,2} \cdot \left( \frac{\sigma_{ss} - \sigma_{ss}}{2} \right) \]
\[ F_N + 2 \cdot \pi \cdot r_{1,2} \cdot \left( \frac{\sigma_{ss} - \sigma_{ss}}{2} \right) = \frac{2}{3} \cdot E^* \cdot \sqrt{r_{1,2} \cdot h_k^3} \] |
<p>| Molerus [18, 19] 1975/78 | Perfect plastic behaviour, varying van der Waals adhesion, shearing | [ F_{H} = F_{H0} + \frac{p_{vaw}}{p_f} \cdot F_N = F_{H0} + \kappa_p \cdot F_N \quad \text{slip at } \sum F_N \leq \mu_i = \tan \phi_i ] |</p>
<table>
<thead>
<tr>
<th>Authors</th>
<th>Description</th>
<th>Formula/Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Schubert et al.</td>
<td>elastic-plastic behaviour, varying van der Waals adhesion force ( F_H )</td>
<td>( F_H = F_{H0} + \frac{p_{VdW}}{p_f} \left( 1 + \frac{2 \cdot A_{el}}{3 \cdot A_{pl}} \right) \cdot F_N ) with ( p_{VdW} = \frac{C_H}{6 \cdot \pi \cdot a_0^3} )</td>
</tr>
<tr>
<td>Rumpf et al.</td>
<td>viscoelastic behaviour, normal force</td>
<td>( r_k^3 = \frac{3 \cdot n_{2,12} \cdot F_N}{2} \left[ \frac{1}{F_0'(t = 0)} + \frac{t}{\eta_k} \right] )</td>
</tr>
<tr>
<td>Rumpf et al.</td>
<td>viscoplastic flow, sintering</td>
<td>( \left( \frac{r_k}{d} \right)^2 = \frac{2 \cdot t}{5 \cdot \eta_k} \left( \frac{2 \cdot \sigma_{ss}}{d} + \frac{1 \cdot F_N}{\pi \cdot d^2} \right) )</td>
</tr>
<tr>
<td>Stieß [167] 1976</td>
<td>high normal load, linear elastic, plastic, viscous damping</td>
<td>( F_N = F_{N,el} + F_{N,pl} + F_{N,vis} = k_{N,tot} \cdot s + F_{N,f} + b \cdot s \left[ 1 - \exp \left( -\frac{k_{N,vis}}{b} \cdot \frac{s}{s_1} \right) \right] ) with ( b = \eta_p \cdot A_0 / d_0 ) ( G(t) = G_0(t_0) \cdot \left{ 1 - \frac{k_{N,vis}}{k_{N, tot}} \left[ 1 - \exp \left( -\frac{k_{N,vis}}{b} \cdot (t - t_0) \right) \right] \right} )</td>
</tr>
<tr>
<td>Savkoor and Briggs</td>
<td>elastic behaviour, constant adhesion, tangential force</td>
<td>( r_k^3 = \frac{3 \cdot n_{1,2} \cdot \left( F_N + F_{HJKR} + \sqrt{2 \cdot F_{HJKR} \cdot F_N + \frac{F_{HJKR}^2}{4}} \right)}{2 \cdot E} )</td>
</tr>
<tr>
<td>Authors</td>
<td>Description</td>
<td>Equation</td>
</tr>
<tr>
<td>-------------------------</td>
<td>-----------------------------------------------------------------------------</td>
<td>----------</td>
</tr>
<tr>
<td>Cundall and Strack [41]</td>
<td>linear elastic, viscous damping (dashpot), rotation</td>
<td>$F_N = k_N \cdot h_K + c_N \cdot \dot{h}<em>K$, $F_T = k_T \cdot \left[ \delta - (\rho_1 \cdot r_1 + \rho_2 \cdot r_2) \right] + c_T \cdot \dot{\delta}$, $F</em>{T,\text{max}} = \tan \varphi_i \cdot F_N + c$</td>
</tr>
<tr>
<td>Maugis and Pollock [144]</td>
<td>elastic-plastic behaviour, normal force, varying adhesion, load, unload</td>
<td>$F_{N,Z,\text{brittle}} = -2 \cdot \sigma_{ss} \cdot E^* \sqrt{\frac{F_N}{\pi \cdot p_f}}$, $F_{N,Z,\text{ductile}} = -\pi \cdot r_{k,f}^2 \cdot \sigma_f$</td>
</tr>
<tr>
<td>Johnson [54]</td>
<td>Elastic, adhesion, elastic-plastic, plastic indentation, elastic-friction, rolling</td>
<td>first yield: $\frac{p_m}{\sigma_f} = \vartheta_f = 1.1$, elastic-plastic: $1.1 \leq \vartheta \leq 3$</td>
</tr>
<tr>
<td>Walton and Braun [145]</td>
<td>plastic behaviour, normal, tangential force, load, unload</td>
<td>$F_{N,\text{load}} = k_N \cdot h_K$, $k_T = k_{T,0} \cdot \left( 1 - \frac{\pm F_T + F_N^*}{\mu_i \cdot F_N + F_T} \right)^{1/3}$</td>
</tr>
<tr>
<td>Thornton and Yin [131] 1991</td>
<td>Elastic behaviour, constant adhesion, tangential force; load, unload, re-load hysteresis</td>
<td></td>
</tr>
<tr>
<td>-----------------------------</td>
<td>----------------------------------------------------------------------------------</td>
<td></td>
</tr>
<tr>
<td>$F_t = 4 \cdot \psi \cdot G \cdot \sqrt{r_{1,2} \cdot h_k} \cdot \Delta \delta \pm (1 - \psi) \cdot \mu_i \cdot \Delta F_N$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\psi_{load}^3 = 1 - \frac{F_t^{*} + \mu_i \cdot \Delta F_N}{\mu_i \cdot F_N}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\psi_{unload}^3 = 1 - \frac{F_t^{*} - F_t + 2 \cdot \mu_i \cdot \Delta F_N}{2 \cdot \mu_i \cdot F_N}$</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sadd, Tai and Shukla [133] 1993</th>
<th>Elastic behaviour, normal load, hysteresis for unload, re-load, re-unload</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_{N,load} = \alpha_{load} \cdot h_k^p$</td>
<td></td>
</tr>
<tr>
<td>$F_{N,unload} = \alpha_{unload} \cdot h_k^{p+q}$</td>
<td></td>
</tr>
<tr>
<td>$q = (k_{N,unload} \cdot h_{k,max})^2$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Thornton and Ning [151] 1998</th>
<th>Elastic-plastic behaviour, normal force, varying adhesion, unload</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_{N,load} = 2/3 \cdot E \cdot \sqrt{r_{1,2} \cdot h_{K,f}} + \pi \cdot r_{1,2} \cdot (h_k - h_{K,f})$</td>
<td></td>
</tr>
<tr>
<td>$F_{N,unload} = 2/3 \cdot E \cdot \sqrt{r_{1,2} \cdot (h_k - h_{K,U})^3}$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Tomas [152] 1999</th>
<th>Elastic-plastic and viscoplastic behaviours, varying adhesion, normal and shear forces</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_H = F_{H0} + \left( \frac{p_{VdW} / p_f}{2/3 + A_{pl}} - 3 \cdot A_k - p_{VdW} / p_f \right) \cdot \left( F_{H0} + F_N \right)$</td>
<td></td>
</tr>
<tr>
<td>$F_R \cdot \left[ \sin 2\alpha - (1 + (\kappa + \kappa_i) \cdot F_{HR} / F_R) \cdot \tan \phi_{pl} \cdot \cos 2\alpha \right] \leq \tan \phi_{pl}$</td>
<td></td>
</tr>
</tbody>
</table>

---

Mechanics_Particle_Adhesion_full.doc © Mechanics of Particle Adhesion Prof. Dr. J. Tomas 09.01.2006
<table>
<thead>
<tr>
<th>Author(s)</th>
<th>Description</th>
<th>Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vu-Quoc and Zhang [136]</td>
<td>1999 Normal and tangential forces, plastic hardening</td>
<td>[ F_{N,(k)} = F_{N,f} - k_N^* \left( \frac{3 \cdot r_{K,2} \cdot F_{N,(0)}}{2 \cdot E^<em>} \right)^{1/3} + k_N^</em> \cdot \sqrt{r_{K,2} \cdot h_K \cdot [1 + k_c \cdot (F_{N,(0)} - F_{N,f})]} ] with [ r_{K,pl,load} = \frac{F_{N} - F_{N,f}}{k_N} \quad r_{K,pl,U} = \frac{F_{N,U} - F_{N,f}}{k_N} ]</td>
</tr>
<tr>
<td>Mesarovic and Johnson [156]</td>
<td>2000 Normal force, elastic, perfectly plastic, elastic-plastic, adhesion, load, unload</td>
<td>[ F_{N,f} = \pi \cdot r \cdot p_f \cdot h_{K,f} \quad \chi = \frac{\sigma_{ss}}{p_f \cdot g(r_{K,f})} = \frac{\pi \cdot \sigma_{ss} \cdot E^*}{4 \pi - 8 \cdot p_f^2 \cdot r_{K,f}} ] detach: [ r_{K,A} = r_{K,f} \cdot \frac{1}{\sqrt{\frac{9}{4} - (\chi - 2) \cdot \chi^3}} ] without adhesion [ F_{N,unload} = \frac{2}{\pi} \left[ \arcsin(x) - x \cdot \sqrt{1 - x^2} \right] ] with [ x = r_K / r_{K,f} ]</td>
</tr>
<tr>
<td>Stronge [55] 2000</td>
<td>Normal force, elastic, elastic-plastic, fully plastic, load, unload</td>
<td>elastic: [ h_K / h_{K,f} = \frac{r_{K,2}^2}{r_{K,f}^2} \quad F_{N,unload} = \frac{2}{3} \cdot E^* \cdot \left( \frac{r_{K,2,U}}{r_{K,f}} \cdot (h_K - h_{K,E}) \right)^3 ] elastic-plastic: [ h_K / h_{K,f} = \frac{1}{2} \left( 1 + \frac{r_{K,2}^2}{r_{K,f}^2} \right) ] fully plastic for [ \frac{h_K}{h_{K,f}} &gt; 84 \quad \frac{r_K}{r_{K,f}} &gt; 12.9 ]</td>
</tr>
<tr>
<td>Tavares and King [134]</td>
<td>2002 n multiple impacts, elastic stiffness reduction by damage</td>
<td>[ F_N = \frac{2}{3} \cdot E^* \cdot (1 - D_n) \cdot \sqrt{r_{K,2} \cdot s_n^3} ] with [ k_{N,n} = k_{N,n-1} \cdot (1 - D_n) ] [ D_n = \left( \frac{S_n}{S_B} \right)^{\gamma_D} ]</td>
</tr>
<tr>
<td><strong>Delenne et al.</strong> [159] 2004</td>
<td>solid bridge bond, flow rule by elliptic paraboloid</td>
<td></td>
</tr>
<tr>
<td>-----------------------------</td>
<td>--------------------------------------------------</td>
<td></td>
</tr>
<tr>
<td>$\zeta = \left( \frac{F_T}{F_{T,f}} \right)^2 + \left( \frac{M_T}{M_{T,f}} \right)^2 - \frac{F_N}{F_{N,Z}} - 1 \leq 0$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>for $\zeta &lt; 0$: $F_N = k_N \cdot h_K$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$F_T = \min(\eta_T \cdot v_T \cdot \mu_i \cdot F_N) \cdot \text{sgn}(v_T)$</td>
<td>$M_T = F_T \cdot r$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th><strong>Gilabert et al.</strong> [234] 2005</th>
<th>normal and tangential forces, constant adhesion, rolling friction</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_N = -F_{H0} \cdot \left[ 1 + \frac{h_K}{h_{K0}} \cdot \Theta(h_K) \right] \cdot \Theta(-h_{K0} - h_K), \quad \Theta(h_K \leq 0) = 0$</td>
<td></td>
</tr>
<tr>
<td>$F_T \leq \mu_i \cdot (F_N + F_{H0})$</td>
<td></td>
</tr>
<tr>
<td>rolling: $F_{R,max} \cdot r = \mu_R \cdot F_N$</td>
<td>$\mu_R &lt; \mu_i \cdot h_i$</td>
</tr>
</tbody>
</table>

### Diagrams
- Solid bridge bond and flow rule by elliptic paraboloid
- Normal and tangential forces, constant adhesion, rolling friction
Fig. 5: Constitutive models of contact deformation of smooth spherical particles in normal direction without (only compression +) and with adhesion (tension -).

The basic models for elastic behaviour were derived by Hertz [108], Fig. 5a), for viscoelasticity by Pao [162], Lee [163], Hunter [164] and Yang [165], Fig. 5c), and for constant adhesion by Johnson et al. [126] and Derjaguin et al. [128], Fig. 5a). Plastic behaviour was described by Stieß [167], Schönert [198], Thornton and Ning [151] and Walton and Braun [145]. But the increase of adhesion force due to plastic contact deformation was introduced by Molerus [18] and Schubert et al. [143], Fig. 5b). Nonlinear plastic, displacement-driven contact hardening was investigated by Vu-Quoc [136], Fig. 5c). Additionally, contact softening could be included [178], Fig. 5c). Viscoelasticity and relaxation was considered by Yang [165], Fig. 5d). Energy dissipation of the
nonlinear elastic contact with viscous spring-dashpot behaviour was modelled by Kuwabara [192], Fig. 5e), during one unload/reload cycle by Sadd et al. [133], Fig. 5f), and for multiple cycles with reduction of their stiffness by Tavares and King [134]. Different elastic, elastic-plastic and fully plastic behaviours were recently described by Stronge [55], Fig. 5h). Time dependent viscoplasticity was modelled by Rumpf et al. [166] and Stieß [167], Fig. 5g). Considering these theories and constitutive models, one obtains a general contact model for load, time and rate dependent viscoelastic, plastic, viscoplastic, adhesion and dissipative behaviours, Tomas [152 - 179], Fig. 5i) and j).

![Constitutive models of contact displacement of smooth spherical particles in tangential direction](image)

Fig. 6: Constitutive models of contact displacement of smooth spherical particles in tangential direction (- sign means reverse shear and displacement directions).

Besides the linear elastic tangential force-displacement relation, Hook’s law Fig. 6 panel a), Fromm [113], Cattaneo [114], Föppl [115], Mindlin [117] and Sonntag [187] described the non-linear contact loading path up to Coulomb friction as the limit, Fig. 6 panel b). The contribution of adhesion forces in Coulomb friction was considered by Derjaguin [128] and Thornton [132], Fig. 6 panel b). Mindlin and Deresiewicz [118], Walton and Braun [145] and Thornton [132]
modelled different non-linear paths for load, unload, reload, reverse shear load, unload and load Fig. 6 panel c). A combination of elastic-frictional and viscous sliding during loading is shown in Fig. 6 panel d). The effect of history or load dependent adhesion $F_H(F_N)$ in Coulomb friction, see Molerus [18 - 21] and Tomas [98, 152] is demonstrated in Fig. 6 panel e) and explained in section 3.7.4.

How to combine these effects as comfortable as possible by applying the continuum methods in microscale should be explained here in detail. This chapter 3 is intended to focus on a characteristic, soft contact of two isotropic, stiff, linear elastic, smooth, mono-disperse spherical particles. This soft or compliant contact displacement is assumed to be small $h_K/d << 1$ compared to the diameter of the stiff particles. The contact area consists of a representative number of molecules. Hence, continuum approaches are only used here to describe the force-displacement behaviour in terms of micromechanics. The microscopic particle shape remains invariant during the dynamic stressing and contact deformation at this nanoscale. In powder processing, these particles are manufactured from uniform material in the bulk phase. These prerequisites are assumed to be suitable for the mechanics of dry nanoparticle contacts in many cases of industrial practice.

### 3.2 Elastic, plastic and viscoplastic contact deformations

#### 3.2.1 Elastic normal force-displacement function

For a single elastic contact of two smooth spheres 1 and 2 with a maximum contact circle radius $r_{K,el}$ but small compared with the particle diameter $d_1$ or $d_2$, an elliptic pressure distribution $p_{el}(r_K)$ is assumed, Hertz [108]:

$$
\left( \frac{p_{el}}{p_{max}} \right)^2 = 1 - \left( \frac{r_K}{r_{K,el}} \right)^2
$$  \hspace{1cm} (1)

With the correlation between compressive repulsion force $F_N$ on each sphere and the maximum pressure $p(r_K = 0) = p_{max}$ in the centre of contact circle at depth $z = 0$,

$$
F_N = 2\pi \int_0^{r_{K,el}} p(r_K) \cdot r_K \, dr_K = \frac{2}{3} \cdot \pi \cdot r_{K,el}^2 \cdot p_{max}
$$  \hspace{1cm} (2)

and the median particle radius $r_{1,2}$ (characteristic radius of contact surface curvature), Fig. 7,

$$
 r_{1,2} = \left( \frac{1}{r_1} + \frac{1}{r_2} \right)^{-1}
$$  \hspace{1cm} (3)

and the average material stiffness as series of elastic elements 1 and 2 (which is equivalent to the sum of element compliances, $E$ modulus of elasticity, $\nu$ Poisson ratio)

$$
E^* = 2 \cdot \left( \frac{1-v_1^2}{E_1} + \frac{1-v_2^2}{E_2} \right)^{-1}
$$  \hspace{1cm} (4)

one can calculate the correlation between normal force $F_N$ and maximum contact radius $r_{K,el}$:
Considering the contributions of an ellipsoidal elastic deformation of the hemispheres \([4, 199]\), contact deformation and surface displacement out of the contact zone, details in Huber \([109]\), the so-called particle centre approach or total overlap height \(h_K\) or normal strain of both particles 1 and 2 is \([108]\):

\[
h_K = h_{K,1} + h_{K,2} = \frac{r_{K,el}^2}{r_{1,2}}
\]

Substitution of Eq. (6) in Eq. (5), results in a non-linear relation between elastic contact force and deformation \([108]\), Fig. 7:

\[
F_N = \frac{2}{3}E^*\sqrt{r_{1,2} \cdot h_K^3}
\]

Eq. (7) is shown as the curve marked Hertz in Fig. 7. The maximum pressure \(p_{\text{max}}\), Eq. (2),

\[
p_{\text{max}} = \frac{3}{2} \cdot \frac{F_N}{\pi \cdot r_{K,el}^2} = \frac{3}{2} \cdot p_m
\]
is 1.5 times the average pressure $p_m$ on the contact area and lies below the micro-yield strength $p_f$. Because of surface bending and, consequently, unconfined yield at the surface perimeter outside of the contact circle $r_K \geq r_{K,el}$, Fig. 7b), a maximum tensile stress is found \[\sigma_{t,max} \approx -0.15 \cdot p_{max}\] (here negative because of positive pressures in powder mechanics):

$$\frac{\sigma_t}{p_{max}} \bigg|_{r_K \geq r_{K,el}} = \frac{1 - 2 \cdot \nu \cdot r_K^2}{3 \cdot r_{K,el}^2}$$

This critical stress for cracking of a brittle particle material with low tensile strength is smaller than the maximum shear stress $\tau_{max} \approx 0.31 \cdot p_{max}$ according to Eq. (12) which is found at the top of a virtual stressing cone below the contact area on the principal axis $r_K^2 = x^2 + y^2 = 0$ in the depth of $z \approx r_{K,el}/2$. Combining the major principal stress distribution, Eq. (10), $\sigma_1 = \sigma_x(z)$ at contact radius $r_K = 0$

$$\frac{\sigma_x(z)}{p_{max}} \bigg|_{r_K \leq r_{K,el}} = \frac{r_{K,el}^2}{r_{K,el}^2 + z^2},$$

and the minor principal stress, Eq. (11), $\sigma_2 = \sigma_y = \sigma(z)$ [109]

$$\frac{\sigma_t(z)}{p_{max}} \bigg|_{r_K \leq r_{K,el}} = -\frac{1}{2} \cdot \frac{r_{K,el}^2}{r_{K,el}^2 + z^2} \cdot \left(1 + \nu \right) \cdot \left[1 - \frac{z}{r_{K,el}} \cdot \arctan \frac{r_{K,el}}{z}\right],$$

the maximum shear stress inside a particle contact $r_K \leq r_{K,el}$ is obtained using the Tresca hypothesis for plastic failure $\tau_{max} = (\sigma_1 - \sigma_2)/2$ [2]:

$$\frac{\tau(z)}{p_{max}} \bigg|_{r_K \leq r_{K,el}} = \frac{3}{4} \cdot \frac{r_{K,el}^2}{r_{K,el}^2 + z^2} \cdot \left[1 - \frac{z}{r_{K,el}} \cdot \arctan \frac{r_{K,el}}{z}\right]$$

This internal shear stress distribution, compare with Lurje [123], becomes more and more critical for ductile or soft solids with a small transition to yield point, and consequently, plastic contact deformation like ultrafine particles with very low contact stiffness, see section 3.4.

The contact stiffness in normal direction is calculated by the first derivative of the force-displacement function (the reciprocal derivative $dh_K/dF_N$ is the compliance):

$$k_N = \frac{dF_N}{dh_K} = E^* \sqrt{r_{1,2} \cdot h_K} = E^* \cdot r_K$$

Due to the parabolic curvature $F_N(h_K)$, the particle contact becomes stiffer with increasing particle size (curve radius of undeformed contact) $r_{1,2}$, contact radius $r_K$ or displacement $h_K$.

### 3.2.2 Elastic-frictional tangential force-displacement function

The influence of an elastic tangential force within a normal loaded circular contact area of spheres was considered by Fromm [113], Cattaneo [114], Sonntag [116, 187], Mindlin [117], Mindlin and Deresiewicz [118], Deresiewicz [40], Hamilton [188], Maw et al. [129], Hess [200], Briscoe [201], Tsuji [202], Johnson [203, 204], Vu-Quoc et al. [135 - 138], Pfister and Eberhard
[140] or Di Renzo and Di Maio [141]. About this and loading, unloading and reloading hysteresis effects, one can find detailed discussions by Johnson [54], Thornton [131], Di Renzo and Di Maio [141], see Table 1 and Fig. 6. The tangential contact force-displacement function is according to the theory of Mindlin [117] (δ = δ1 - δ2 relative tangential displacement within the contact plane, µi coefficient of internal friction between particles 1 and 2 in this contact shear plane):

\[
F_T = \mu_1 \cdot F_N \left[ 1 - \left( 1 - \frac{8 \cdot G^* \cdot r_{K,el}}{3 \cdot \mu_1 \cdot F_N} \cdot \delta \right)^{3/2} \right] \tag{14}
\]

\[G_1 = E_1/(1 + v_1)\] is the shear modulus and the averaged shear modulus \(G^*\) is given as:

\[
G^* = 2 \cdot \left( \frac{2 - v_1}{G_1} + \frac{2 - v_2}{G_2} \right)^{-1} \tag{15}
\]

This model considers slip-stick behaviour within the contact plane and the radius of the annular slip region \(r_{K,slip}\) is calculated by superposition of elliptic pressure distributions, Eq. (1), [54]:

\[
F_T = 2 \pi \cdot \mu_1 \cdot p_{\text{max}} \int_0^{r_{K,el}} \left( 1 - \frac{r^2_{K}}{r^2_{K,el}} \right)^{1/2} r^*_K \cdot dr^*_K - 2 \pi \cdot p_{\text{max}} \int_0^{r_{K,slip}} \left( 1 - \frac{r^2_{K}}{r^2_{K,slip}} \right)^{1/2} r^*_K \cdot dr^*_K \tag{16}
\]

\[
r_{K,slip} = \left( 1 - \frac{F_T}{\mu_1 \cdot F_N} \right)^{1/3} \tag{17}
\]

If, keeping \(F_N\) constant, the tangential force \(F_T\) is increased steadily from zero, micro-slip begins immediately at the perimeter of the contact area \(r_{K,slip} = r_{K,el}\) and spreads inwards according to Eq. (17). When \(F_T\) approaches the friction limit \(\mu_i \cdot F_N\), the boundary between stick-slip \(r_{K,slip}\) approaches zero and the circular stick region shrinks to the centre point at \(x = y = 0\) [54]. This completely mobilized frictional contact gliding \(F_{T,\text{max}} = \mu_i \cdot F_N\), i.e. for Coulomb friction, is obtained for the displacement limit \(\delta_C\) in Eq. (14), see sketch in Table 1 left at line Mindlin [117]:

\[
\delta \geq \delta_C = \frac{3 \cdot \mu_1 \cdot F_N}{8 \cdot G^* \cdot r_{K,el}} \tag{18}
\]

The first derivative of the tangential force-displacement curve is the tangential stiffness \(k_T\):

\[
k_T = 4 \cdot G^* \cdot r_{K,el} \cdot \left( 1 - \frac{\delta}{\delta_C} \right)^{1/2} = 4 \cdot G^* \cdot r_{K,el} \cdot \sqrt{1 - \frac{8 \cdot G^* \cdot r_{K,el}}{3 \cdot \mu_1 \cdot F_N} \cdot \delta} \tag{19}
\]

Due to the digressive curvature \(F_T(\delta)\) the tangential contact stiffness decreases with increasing displacement \(\delta\) and becomes zero for \(\delta = \delta_C\) when it reaches the Coulomb friction limit. The initial tangential contact stiffness (at \(\delta = 0\)) results in this simple function:

\[
k_{T,0} = \left. \frac{dF_T}{d\delta} \right|_{\delta = 0} = 4 \cdot G^* \cdot r_K = 4 \cdot G^* \cdot \sqrt{r_{1,2} \cdot h_K} \tag{20}
\]
Equivalent to the normal stiffness $k_N$, Eq. (13), the initial tangential stiffness $k_{T,0}$ becomes larger with increasing particle size (curve radius of virgin contact) $r_{1,2}$, contact radius $r_K$ or normal displacement $h_K$. Thus, the ratio of the initial tangential stiffness to the normal stiffness is:

$$\frac{k_{T,0}}{k_N} = \frac{4 \cdot G \cdot r_K}{E \cdot r_K} = \frac{2 \cdot (1 - \nu)}{2 - \nu}$$

Hence this ratio ranges from unity, for $\nu = 0$, to $2/3$, for $\nu = 0.5$ [117], which is different from the common linear elastic behaviour of a cylindrical rod.

Mindlin and Deresiewicz [118] supplemented this model by the function for unload and shear in reverse direction ($-$ sign)

$$\delta = \delta_C \cdot \left[ 2 \left( 1 - \frac{F_{T,U}^* - F_T}{2 \cdot \mu_i \cdot F_N} \right)^{2/3} - \left( 1 - \frac{F_{T,U}^*}{\mu_i \cdot F_N} \right)^{2/3} \right],$$

or rearranged as force-displacement relation using Eq. (14) for the unload point $F_{T,U}^* (\delta_U)$:

$$F_{T,U} = F_{T,U}^* - 2 \cdot \mu_i \cdot F_N \cdot \left[ 1 - \left( 1 - \frac{\delta_U - \delta}{2 \cdot \delta_C} \right)^{3/2} \right]$$

Consequently, reload in the previous ($+$) shear direction is for the point $F_{T,\text{reload}} = -F_{T,U}^*$, $\delta_{\text{reload}} = -\delta_U$, see sketch in Table 1 right:

$$F_{T,\text{reload}} = -F_{T,\text{reload}}^* + 2 \cdot \mu_i \cdot F_N \cdot \left[ 1 - \left( 1 - \frac{\delta + \delta_{\text{reload}}}{2 \cdot \delta_C} \right)^{3/2} \right]$$

Thornton has combined these tangential force-displacement relations in one formula [131, 132]

$$F_T = 4 \cdot \psi \cdot G \cdot \sqrt{r_{1,2}} \cdot h_K \cdot \delta \pm \left( 1 - \psi \right) \cdot \mu_i \cdot \Delta F_N,$$

with the loading parameter $\psi$ dependent on loading, unloading and reloading, see in Table 1 Thornton and Yin [131].

Mechanical work is dissipated per one closed cycle (curve integral) between unload $\delta_U$ and reload $-\delta_{\text{reload}}$ displacements according to Eqs. (23) and (24):

$$W_T = \oint_C F_T (\delta) \, d\delta = \int_{-\delta_{\text{reload}}}^{\delta_U} F_{T,\text{reload}} (\delta) \, d\delta - \int_{-\delta_U}^{-\delta_{\text{reload}}} F_{T,U} (\delta) \, d\delta$$

$$W_T = 8 \cdot \mu_i \cdot F_N \cdot \delta_C \cdot \left\{ 1 - \left( \frac{F_{T,\text{reload}}^* + F_{T,U}^*}{4 \cdot \mu_i \cdot F_N} \right)^{5/2} \right\}$$

### 3.2.3 Rolling resistance and elastic-frictional twisting of an elastic particle contact

Besides Coulomb friction, the sources of an additional rolling resistance $F_R$ should be considered by a so-called “microslip”, micro-roughness of contact surfaces, and especially, by contact de-
formations [54, 113, 187, 227 - 234]. The rolling resistance is a moment (torque) \( M_R \) counteracting relative angular velocity of particle \( \omega = d\gamma /dt \). Because of elastic and inelastic contact flattening, it can be explained microscopically by antiparallel normal forces acting on the opposite sides of the contact perimeter with total separation \( 2r_{K,el} \) [52].

Rolling with partial slip up to reaching the Coulomb friction limit was described by a “creep” coefficient, see Johnson [54], for a soft sphere driven by a longitudinal velocity \( v \) and rolling on a relatively rigid plane surface

\[
\xi = \frac{(\omega_f - \omega_s) \cdot r}{v} = \frac{3 \cdot \mu_i \cdot F_N \cdot (4 - 3 \cdot v)}{16 \cdot G \cdot r_{K,el}^2} \cdot \left[ 1 - \left( \frac{F_R}{\mu_i \cdot F_N} \right)^{1/3} \right],
\]

(28)

and for no-slip:

\[
\xi = \frac{(4 - 3 \cdot v)}{16 \cdot G \cdot r_{K,el}^2} \cdot F_R
\]

(29)

Moreover, one can distinguish between the linear elastic contribution of moment-rolling angle relation (\( k_R \) rolling stiffness, \( \alpha_R \) coefficient for friction losses)

\[
M_R = F_R \cdot r = k_R^* \cdot \gamma = \alpha_R \cdot k_R \cdot \gamma
\]

(30)

and a so-called “frictional” contribution which is more or less equivalent to contact sliding. This frictional rolling resistance of smooth soft spheres can be considered by a tilting moment relation of the force pair around the pivot at the perimeter of contact circle, see Fig. 2 panel e). As the response of normal load \( F_N \), contact flattening and a lever arm \( r_{K,el}^* \), the perpendicular rolling resistance \( F_{R,C} \) acts at a lever arm \( r-h_K/2 \). With Eq. (5) for \( r_{K,el}^* \) one obtains:

\[
F_{R,C} = \frac{r_{K,el}^* \cdot F_N}{r - h_K / 2} \approx \frac{r_{K,el}^* \cdot F_N}{r} = \left( \frac{3 \cdot F_N^4}{2 \cdot r_{K,el}^* \cdot E^*} \right)^{1/3}
\]

(31)

This resistance yields the function \( F_R(F_N) \propto F_N^{4/3} \) and is much smaller than the condition for a sliding contact \( F_R < \mu_i \cdot F_N \). There is no rolling resistance for perfectly rigid spheres with point contact \( r_{K,el} = 0 \). The ratio \( r_{K,el}^* / r = \mu_R \approx 10^{-3} - 10^{-1} \) is equivalent to the coefficient of rolling friction used in engineering practice to describe the rolling resistance. For an inelastic adhesive contact this coefficient of rolling friction depends on the load history, explained in section 3.7.5.

Another effect is that the sphere can rotate (twist or spin) around its principal axis (here z-axis) within the contact plane, see 7th panel in Table 1 right. The torque \( M_{lo} \) as radial distribution versus radius coordinate \( r_K^* \) of circular elastic contact area, i.e. for \( 0 \leq r_K^* \leq r_{K,el}^* \),

\[
M_{lo} = 2\pi \int_0^{r_{K,el}^*} \tau(r_K^*) \cdot r_K^{*2} \cdot dr_K
\]

(32)

and as function of the rotation angle \( \phi \) was calculated by Mindlin [117], Cattaneo [120, 121], Lubkin [119], Deresiewicz [40, 122] and Johnson [54]. The elastic-frictional torsional stiffness
$k_{to}$ (resistance) for one particle obtained on the assumption of an annulus of slip (partial slip) results in [40]:

$$k_{to} = \frac{dM_{to}}{d\phi} = \frac{8 \cdot G \cdot r_{el,K}^3}{3} \left[ 2 \cdot \left( 1 - \frac{3 \cdot M_{to}}{2 \cdot \mu_1 \cdot F_N \cdot r_{K,el}} \right)^{-1/2} - 1 \right]^{-1} \tag{33}$$

The initial elastic torsional stiffness $k_{to,0}$ at $M_{to} = 0$ in the absence of slip was first derived by Mindlin [117] (here with $\phi = \phi_1 + \phi_2$ and $G = 2 \cdot (1/G_1 + 1/G_2)^{-1}$ instead of $G^*$, see Eq. (15)):

$$k_{to,0} = \frac{8 \cdot G \cdot r_{el,K}^3}{3} \tag{34}$$

With Eq. (5) for $r_{K,el}$ and Eq. (21) for $G^*/E^*$ this initial torsional stiffness can also be written as:

$$k_{to,0} = 2 \cdot (1 - \nu) \cdot r_{1,2} \cdot F_N \tag{35}$$

Similar to the Coulomb friction limit of tangential displacement $\delta_C$, Eq. (18), completely mobilized frictional contact rotation is obtained at stiffness $k_{to} = 0$ for the moment limit $M_{to,C}$:

$$M_{to} \geq M_{to,C} = \frac{2 \cdot \mu_1 \cdot F_N \cdot r_{K,el}}{3} \tag{36}$$

Additionally, mechanical work is dissipated per one closed twisting cycle of an oscillating moment $\pm M_{to,U}$ [40]:

$$W_{to} = \frac{16 \cdot (\mu_1 \cdot F_N)^2}{9 \cdot G \cdot r_{K,el}} \left[ 1 - \left( 1 - \frac{M_{to,U}}{M_{to,C}} \right)^{3/2} \right] \cdot \frac{3 \cdot M_{to,U}}{4 \cdot M_{to,C}} \cdot \left[ 1 + \left( 1 - \frac{M_{to,U}}{M_{to,C}} \right)^{1/2} \right] \tag{37}$$

Generally, sliding, rolling and twisting are coupled in the moving particle packing, e.g. [52, 235].

### 3.2.4 Elastic displacement of an adhesive contact

The adhesion in the normal loaded contact of spheres with elastic displacement will be additionally shown. For fine and stiff particles, the Derjaguin, Muller and Toporov DMT model [110, 128, 205] predicts that half of the interaction force $F_{H,DMT}/2$ occurs outside in the annular area which is located at the perimeter closed by the contact, Eq. (38). This is in contrast to the Johnson, Kendall and Roberts JKR model [54] which assumes that all the interactions occur within the contact radius of the particles. The median adhesion force $F_{H,DMT}$ of a direct contact of spherical particles can be expressed in terms of the work of adhesion $W_A$, conventional surface energy $\gamma_A$ or surface tension $\sigma_{sls}$ $W_A = 2 \cdot \gamma_A = 2 \cdot \sigma_{sls}$. The index sls means particle surface - adsorption layers (with liquid equivalent mechanical behaviour) – particle surface interaction. If only molecular interactions with separations near the contact contribute to the adhesion force then the so-called Derjaguin approximation [110] is valid

$$F_{H,DMT} = 4 \cdot \pi \cdot \sigma_{sls} \cdot r_{1,2}, \tag{38}$$
which is in agreement with Bradley’s formula [111]. This surface tension $\sigma_{\text{sls}}$ equals half the energy needed to separate two flat surfaces from an equilibrium contact distance $a_{F=0}$ to infinity [58]:

$$\sigma_{\text{sls}} = \frac{1}{2} \cdot \left( \int_{a_{F=0}}^{\infty} p_{\text{VdW}}(a) \, da \right) = \frac{C_{\text{H,sls}}}{24 \cdot \pi \cdot a_{F=0}^2}$$  \hspace{1cm} (39)

The adhesion force per unit planar surface area or attractive pressure $p_{\text{VdW}}$ which is used here to describe the van der Waals interactions at contact is equivalent to a theoretical bond strength and can simply calculated as [58] (e.g. $p_{\text{VdW}} \approx 3 – 600$ MPa):

$$p_{\text{VdW}} = \frac{C_{\text{H,sls}}}{6 \cdot \pi \cdot a_{F=0}^3} = \frac{4 \cdot \sigma_{\text{sls}}}{a_{F=0}^3}$$  \hspace{1cm} (40)

Using this and for comparison of the adhesion or bond strength, a dimensionless ratio of elastic adhesion displacement (elastic extension at contact detachment), expressed as height of “neck” $h_{N,T}$ around the contact zone, to minimum molecular centre separation $a_{F=0}$ can be defined as

$$\Phi_T = h_{N,T}^{-1} = \left( \frac{4 \cdot r_{1,2} \cdot \sigma_{\text{sls}}^2}{E^* \cdot a_{F=0}^3} \right)^{1/3} = \left( \frac{r_{1,2} \cdot p_{\text{VdW}}^2}{4 \cdot E^* \cdot a_{F=0}^3} \right)^{1/3}$$  \hspace{1cm} (41)

which was first introduced by Tabor [208] and later modified and discussed by Muller [205, 206], Maugis [209], Greenwood [147] and Johnson [156]. The DMT model works for very small and stiff particles $\Phi_T < 0.1$ [205, 209, 212]. For separating a stiff, non-deformed spherical point contact, the DMT theory [128] predicts a necessary pull-off force $F_{N,Z}$ equivalent to the adhesion reaction force expressed by Eq. (38).

Compared with the stronger covalent, ionic, metallic or hydrogen bonds, these particle interactions are comparatively weak. From Eq. (39) the surface tension is about $\sigma_{\text{sls}} = 0.25 – 50$ mJ/m$^2$ or the Hamaker constant according to Lifshitz continuum theory amounts to $C_{\text{H,sls}} = (0.2 – 40) \cdot 10^{-20}$ J [58, 210, 211]. Notice here, the particle interactions depend greatly on the applied load, which is experimentally confirmed by atomic force microscopy [214, 216, 217].

A balance of stored elastic energy, mechanical potential energy and surface energy delivers the contact radius of two spheres [126], expressed here with a constant adhesion force $F_{H,JKR}$ regarding Eq. (44):

$$r_k^3 = \frac{3 \cdot r_{1,2}^2}{2 \cdot E^*} \left( F_N + F_{H,JKR} + \sqrt{2 \cdot F_{H,JKR} \cdot F_N + F_{H,JKR}^2} \right)$$  \hspace{1cm} (42)

Eq. (42) indicates a contact radius increase with increasing work of adhesion. The contact force-displacement relation is obtained from Eqs. (6) and (42) and can be compared with the Hertz relation Eq. (7) by the curves in Fig. 5a) marked with Hertz and JKR:

$$F_N = \frac{2 \cdot E^* \cdot r_{1,2} \cdot h_k^3}{3} - \sqrt{\frac{4 \cdot E^* \cdot F_{H,JKR}}{3} \cdot \frac{r_{1,2} \cdot h_k^3}{3}}$$  \hspace{1cm} (43)
For small contact deformation the so-called JKR limit [126] is half of constant adhesion force $F_{H,JKR}$. This JKR model can be applied for higher bond strength $\Phi_T > 5$ [209, 212, 213, 218]. It is valid for comparatively larger and softer particles than the DMT model predicts:

$$F_{H,JKR} = \frac{F_{H,JKR}}{2} = 3 \cdot \pi \cdot \sigma_{sl} \cdot r_{1,2}$$  \hspace{1cm} (44)

Thus the contact radius for zero load $F_N = 0$,

$$r_{K,0} = \sqrt{\frac{3 \cdot r_{1,2} \cdot F_{H,JKR}}{E^*}}$$  \hspace{1cm} (45)

is reduced to the pull-off contact radius, i.e.,

$$r_{K,pull-off} = r_{K,0}/\sqrt{4}$$  \hspace{1cm} (46)

Johnson [204] extended his JKR model by an adhesion force contribution outside in the annular radius $r_{K,c} \geq r_{K,el}$ which is located at the perimeter closed by the contact (zone of surface bending). Using the Maugis-Dugdale model [209] he obtained with $x = r_{K,c}/r_K$:

$$F_N = 4 \pi \cdot \sigma_{sl} \cdot r_{1,2} \left( \frac{2 \cdot E^* \cdot r_{K}^3}{3 \pi \cdot \sigma_{sl} \cdot r_{1,2}^2} - 1.16 \cdot \Phi_T \cdot \left( \frac{2 \cdot E^* \cdot r_{K}^3}{3 \pi \cdot \sigma_{sl} \cdot r_{1,2}^2} \right)^{2/3} \sqrt{x^2 - 1 + x^2 \cdot \arccos \left( \frac{1}{x} \right)} \right)$$  \hspace{1cm} (47)

Additionally, with applying an increasing tangential force $F_T$, the contact radius $r_K$ is reduced by the last term within the square root:

$$r_{K}^3 = \frac{3 \cdot r_{1,2}^{2} \left( F_N + F_{H,JKR} + \sqrt{2 \cdot F_{H,JKR} \cdot F_N + F_{H,JKR}^2 - \frac{F_T^2 \cdot E^*}{4 \cdot G^*}} \right)^2}{2 \cdot E^*}$$  \hspace{1cm} (48)

When the square root in Eq. (48) disappears to zero, a critical value $F_{T,crit}$ is obtained, the so-called “peeling” of contact surfaces [189, 190]:

$$F_{T,crit} = 2 \cdot \sqrt{\frac{2 \cdot F_{H,JKR} \cdot F_N + F_{H,JKR}^2}{E^*} \cdot G^*}$$  \hspace{1cm} (49)

An effective or net normal force ($F_N + F_{H,JKR}$) remains additionally in the contact [132]. Considering $F_T > F_{T,crit}$, i.e., contact failure by sliding, see Mindlin [118], the tangential force limit is expressed as $F_T = \tan \varphi_i \cdot (F_N + F_{H0})$. The adhesion force $F_{H0}$ (index H0) is constant during contact failure and the coefficient (or angle) of internal friction $\mu_i = \tan \varphi_i$ is also assumed to be constant for a multi-asperity contact [125, 212, 220]. This constant friction was often confirmed for rough surfaces in both elastic and plastic regime, e.g. [125, 218, 220], but not for a single-asperity contact with nonlinear dependence of friction force on normal load [212, 218].

Rearranging Eq. (48), the extended contact force-displacement relation shows a reduction of the Hertz (first square-root) and JKR contributions to normal load $F_N$ which is needed to obtain a given displacement $h_K$:

$$F_N = \frac{2 \cdot E^*}{3} \cdot \sqrt{r_{1,2} \cdot h_K^3} - \sqrt{\frac{4 \cdot E^* \cdot F_{H,JKR}}{3} \cdot \sqrt{r_{1,2} \cdot h_K^3} - \frac{F_T^2 \cdot E^*}{4 \cdot G^*}}$$  \hspace{1cm} (50)
However in terms of ultrafine particles \( d < 10 \, \mu \text{m} \), the increase of contact area with elastic deformation does not lead to a significant increase of attractive adhesion forces because of a too small van der Waals energy of adhesion, Eq. (39). The reversible elastic repulsion restitutes always the initial contact configuration during unloading. Consequently, the increase of adhesion by compression, e.g. forming a snow ball, the well-known cohesive consolidation of a powder or the particle interaction and remaining strength after tabletting must be influenced by irreversible contact deformations, which are shown for a small stress level in Fig. 8.

Fig. 8: Representative particle contact deformation – c) elastic-plastic compression (limestone, median particle size \( d_{50,3} = 1.2 \, \mu \text{m} \)) [155]. The dominant linear elastic-plastic deformation range between macroscopic stress levels of powder mechanics \( \sigma = 1 - 50 \, \text{kPa} \) [7] is demonstrated here. If the maximum pressure in the contact centre reaches the micro-yield strength \( p_f = 300 \, \text{N/mm}^2 \) at the yield point \( Y \) (\( F_{N,f} = -0.74 \, \text{nN} \) for \( E = 150 \, \text{kN/mm}^2 \), \( \nu = 0.28 \)) then the contact starts with plastic yielding which is intensified by surface defects and/or adsorption layers. The combined elastic-plastic yield limit of the partial plate-plate contact is achieved as given in Eq. (99).

If the maximum pressure \( p_{\text{max}} \) in the centre of the contact circle reaches the micro-yield strength \( p_f \), the contact starts with irreversible plastic yielding (index \( f \)). From Eqs. (2) and (5) these elastic limits of radius \( r_{K,f} \) and from Eq. (6) of the centre approach \( h_{K,f} \) are calculated as:

\[
r_{K,f} = \frac{\pi \cdot r_{1,2} \cdot p_f}{E^*} \tag{51}
\]

\[
h_{K,f} = r_{1,2} \cdot \left( \frac{\pi \cdot p_f}{E^*} \right)^2 \tag{52}
\]

Fig. 8 demonstrates the dominant irreversible contact deformation over a wide range of normal forces \( F_N >> F_{N,f} = -0.74 \, \text{nN} \).
The micro-yield strength \( p_f \), especially, of ultrafine inorganic/organic powders from processing plants is normally unknown. This constitutive value is significantly influenced by surface micro-properties of the particles, e.g. by surface defects, nano-asperities and immobile/mobile adsorption layers. In terms of known material strength properties, e.g. for coarse particles made by metals, one may use the flow rule according to Tresca, Johnson [54], or the von Mises, Huber and Hencky yield criterion [161]. Vu-Quoc et al. [136 - 139] explain the equivalent stress \( p_f \) by the continuum theory of plasticity, i.e. by the second invariant of stress deviator. But if one considers a confined plastic stress field in contact flattening and indentation, equivalent to a soft deformation zone within a stiff “shell”, a simple relation is here with material’s uniaxial yield stress \( \sigma_f \) for tension:

\[
\frac{2}{3} \cdot p_{\text{max}} = p_m \approx (1.1 - 2.8) \cdot \sigma_f
\]  

Between \( 1.1 \leq \vartheta = p_m / \sigma_f \leq 2.8 \) an increasing average value of the elastic-plastic pressure \( p_m \) at increasing contact radius \( r_k \) can also be calculated from the radial stress distribution \( \sigma_r(r_k) \) which is located around the principal z-axis beneath a smooth sphere indenting a flat surface hold in a fixed position [54]:

\[
\frac{p_m}{p_f} = \frac{\sigma_r(r_k)}{p_f} = 1 + \frac{2 \cdot \sigma_f}{3 \cdot p_f} \cdot \ln \frac{r_k}{r_{k,\text{f}}}
\]

A factor \( \vartheta = 3 \) is recommended in terms of contact plasticity with hardening, e.g. for ductile alloys [136]. But for inorganic or mineral particles with high macroscopic compressive strength but low tensile strength, this microscopic yield strength in compression (micro-hardness) \( p_f \approx (3 - 15) \cdot \sigma_f \) can roughly be correlated by the yield strength under uniaxial tension \( \sigma_f \), see Fischer-Cripps [5], Gane [221] or Ghadiri [222].

Rumpf et al. [166] and Molerus [18, 19] introduced their contact models in powder mechanics which are used in particle technology. The JKR theory was the basis of adhesion mechanics [54, 59, 144, 156, 204, 209, 223 - 226] and was also set in analogy to fracture mechanics [59, 148, 223] with respect to fracture toughness and crack propagation in solid materials.

### 3.2.5 Perfect plastic and viscoplastic contact displacement and viscous damping

Actually, assuming perfect contact plasticity one can neglect the surface deformation outside of the contact zone. With the following geometrical relation of a sphere

\[
r_k^2 = r_1^2 - (r_1 - h_{k,1})^2 = 2 \cdot r_1 \cdot h_{k,1} - h_{k,2}^2 \approx d_1 \cdot h_{k,1}
\]

one obtains the total particle centre approach of both spheres as series of two elements approaches:

\[
h_k = h_{k,1} + h_{k,2} = \frac{r_k^2}{d_1} + \frac{r_k^2}{d_2} = \frac{r_k^2}{2 \cdot r_{l,2}}
\]
Because of this, a linear force-displacement relation is found for a small contact between spherical particles. The repulsive force as a resistance against plastic deformation is given as:

$$F_{N,pl} = p_f \cdot A_k = \pi \cdot d_{1,2} \cdot p_f \cdot h_k$$  \hspace{1cm} (57)

Thus, the contact stiffness is constant for perfect plastic yielding behaviour, but decreases with smaller particle diameter $d_{1,2}$ especially for cohesive ultrafine particles:

$$k_{N,pl} = \frac{dF_N}{dh_k} = \pi \cdot d_{1,2} \cdot p_f$$  \hspace{1cm} (58)

Additionally as a first approach, the rate dependent, perfect viscoplastic deformation (at the point of yielding) expressed by apparent contact viscosity $\eta_k$ times indentation rate $h_k$ is assumed to be equivalent to yield strength $p_f$ multiplied by indentation height increment $h_k$:

$$p_f \cdot h_k = \eta_k \cdot h_k,$$  \hspace{1cm} (59)

and one obtains again a linear model regarding strain rate:

$$F_{N,vis} = \eta_k \cdot \dot{A}_k = \pi \cdot d_{1,2} \cdot \eta_k \cdot \dot{h}_k$$  \hspace{1cm} (60)

An attractive viscous force is observed, e.g. for capillary numbers $Ca = \eta_k \cdot \dot{h}_k / \sigma_{lg} > 1$ when comparatively strong bonds of (low-viscous) liquid bridges are extended with negative velocity $-\dot{h}_k$ [102 - 104].

Besides the linear spring-dashpot model [41] in discrete element modelling (DEM) viscous damping is introduced as parallel arrangement of an elastic conservative force due to the deformation $h_k$ and a viscous force depending on the deformation rate $\dot{h}_k$ [44, 192, 193]:

$$F_N = F_{N,el} + F_{N,vis} = E \left( \frac{2}{3} \cdot \sqrt{r_{1,2} \cdot h_k^3} + C_{vis} \cdot \sqrt{r_{1,2} \cdot h_k \cdot \dot{h}_k} \right)$$  \hspace{1cm} (61)

This viscous force $F_{N,vis}$ is weighted by the displacement $h_k$ [44] and $C_{vis}$ is a dissipative constant which characterises the viscous stiffness or the apparent contact viscosity [192, 193]:

$$\eta_k = C_{vis} \cdot E \cdot \sqrt{r_{1,2} \cdot h_k}$$  \hspace{1cm} (62)

Consequently, the particle material parameters contact micro-yield strength $p_f$ and contact viscosity $\eta_k$ are measures of irreversible particle contact stiffness or softness. Both plastic and viscous contact yield effects were intensified by surface defects and mobile adsorption layers. The sum of deformation increments results in the energy dissipation.

For high normal loads $F_N$ applied in compressive stressing in comminution or press agglomeration processes, large particle contact areas $A_k$ and particle volume deformation, as response, are expected. This can be modelled by the conventional linear elastic and constant plastic behaviour and viscous damping. Thus, a rheological model as parallel sequence of the linear spring (Hook model, $k_{N,el}$ stiffness), the plasticity (Prandtl model, $F_{N,pl}$ plastic yield limit) and the dashpot (Maxwell model, $\eta_p$ particle or solid viscosity) was used by Steß [167]. He describes the force-displacement relation ($A_0$ initial cross-sectional area, $d_0$ initial particle diameter)
with the total displacement, i.e. the sum of the contact displacement and a contribution by particle deformation, \( s \geq s_f = \frac{F_{N,f}}{k_{N,pl}} \) for yield. Every parallel elastic, plastic and viscous force contribution has implemented a sequential spring stiffness \( k_{N,el}, k_{N,pl} \) and \( k_{N,vis} \). Thus, the initial slope of the force-displacement curve is for \( t = 0 \),

\[
\frac{dF_N}{ds}igr|_{t=0} = k_{N,tot} = k_{N,el} + k_{N,pl} + k_{N,vis}
\]

and for a comparatively large time \( t > 3 \cdot t_{63,vis} = 3 \cdot \eta_p \cdot A_0 / (k_{N,vis} \cdot d_0) \) one obtains [167]:

\[
F_N(t \to 3 \cdot t_{63,vis}) = k_{N,el} \cdot s + F_{N,f} + \eta_p \cdot A_0 \cdot \dot{s} / d_0
\]

Comparing the coefficients of the last term of Eq. (65) to Eq. (60) one can also write for a viscoplastic particle contact with a characteristic time parameter \( t_{63,K} = \eta_k / p_f \):

\[
F_{N,vis} = \pi \cdot d_{1,2} \cdot \eta_k \cdot h_k \cdot \left[ 1 - \exp\left( -\frac{t}{t_{63,K}} \right) \right]
\]

Recently, contact viscoelasticity and constant adhesion force on basis of JKR model were combined by analysing the elastic and dissipative stress tensors, Brilliantov and Pöschel [171, 172]. Particle swelling and shrinkage are modelled by a rate function \( \dot{s} \propto \dot{d} / d \) [238].

Taking into consideration these facts, what are the consequences of small contact flattening with respect to a varying, i.e., load or pre-history dependent adhesion?

### 3.3 Load dependent adhesion force

#### 3.3.1 Increase of adhesion force by contact consolidation

Krupp [57] and Sperling [124, 142] developed a model for the increase of the total adhesion force \( F_H \) (index \( H \), positive sign) of the contact. This considerable effect is called here as “consolidation” and is expressed as the sum of adhesion force \( F_{H0} \) according to Eq. (38) and an attractive/repulsive force contribution due to irreversible plastic flattening of the spheres (\( p_f \) is the repulsive “microhardness” or micro-yield strength of the softer contact material of both particles, \( \sigma_{ss}/a_0 \) is the attractive contact pressure, index \( ss \) represents direct solid-vacuum-solid interaction):

\[
F_H = 4 \cdot \pi \cdot r_{1,2} \cdot \sigma_{ss} \cdot \left( 1 + \frac{2 \cdot \sigma_{ss}}{a_0 \cdot p_f} \right)
\]

Dahneke [127] modified this adhesion model by the van der Waals force without any contact deformation \( F_{H0} \) plus an attractive van der Waals pressure (force per unit surface) \( p_{VdW} \) contribution due to partially increasing flattening of the spheres which form a circular contact area \( A_K \) (\( C_H \) is the Hamaker constant based on interacting molecule pair additivity [58, 210]):

\[
F_H = F_{H0} + A_K \cdot p_{VdW} = \frac{C_H \cdot r_{1,2}}{6 \cdot a_0^2} \cdot \left( 1 + \frac{2 \cdot h_K}{a_0} \right)
\]
The distance $a_0$ denotes a characteristic adhesion separation. If stiff molecular interactions provided (no compression of electron sheath), this molecular centre separation $a_0$ was assumed to be constant during contact loading. By addition the elastic repulsion of the solid material according to Hertz, Eq. (7), to this attraction force, Eq. (68), and by deriving the total force $F_{tot}$ with respect to $h_K$, the maximum adhesion force was obtained as absolute value

$$F_{H,max} = \frac{C_H \cdot r_{1,2}}{6 \cdot a_0^2} \left( 1 + \frac{2 \cdot C_H^2 \cdot r_{1,2}^2}{27 \cdot E_s^2 \cdot a_0^2} \right),$$

(69)

which occurs at the centre approach of the spheres [127]:

$$h_{K,max} = \frac{C_H^2 \cdot r_{1,2}}{9 \cdot E_s^2 \cdot a_0^6}$$

(70)

But as mentioned before, this increase of contact area with elastic deformation does not lead to a significant increase of attractive adhesion force. The reversible elastic repulsion restitutes always the initial contact configuration. The practical experience with the mechanical behaviour of fine powders shows that an increase of adhesion force has to be influenced by an irreversible or “frozen” contact flattening which depends on the external force $F_N$ [143].

Generally, if this external compressive normal force $F_N$ is acting at a single soft contact of two isotropic, stiff, smooth, mono-disperse spheres the previous contact point is deformed to a contact area, Fig. 7a) and Fig. 8c), and the adhesion force between these two partners increases, see in Fig. 10 the so-called “adhesion limit” for incipient contact detachment. During this surface stressing the rigid particle is not so much deformed that it undergoes a certain change of the particle shape. In contrast, soft particle matter such as biological cells or macromolecular organic material does not behave so.

### 3.3.2 Linear increase of adhesion force by normal force

For soft contacts Rumpf et al. [166] have developed a constitutive model approach to describe the linear increase of adhesion force $F_H$, mainly for plastic contact deformation:

$$F_H = \left( 1 + \frac{p_{VdW}}{p_f} \right) \cdot F_{H0} + \frac{p_{VdW}}{p_f} \cdot F_N = \left( 1 + \kappa_p \right) \cdot F_{H0} + \kappa_p \cdot F_N$$

(71)

With analogous prerequisites and derivation, Molerus [19] obtained a similar expression:

$$F_H = F_{H0} + \frac{p_{VdW}}{p_f} \cdot F_N = F_{H0} + \kappa_p \cdot F_N$$

(72)

The adhesion force $F_{H0}$ without additional consolidation ($F_N = 0$) can be approached as a single rigid sphere-sphere contact, Fig. 7a). But, if this particle contact is soft enough the contact is flattened by an external normal force $F_N$ to a plate-plate contact, Fig. 8c). The coefficient $\kappa_p$ describes a dimensionless ratio of attractive van der Waals pressure $p_{VdW}$ for a plate-plate model, Eq. (40), to repulsive particle micro-hardness $p_f$ which is temperature sensitive.
\[ \kappa_p = \frac{p_{\text{vdW}}}{p_f} = \frac{C_{H,\text{sls}}}{a_{F=0} \cdot p_f} = 4 \cdot \frac{\sigma_{\text{sls}}}{a_{F=0} \cdot p_f} \]

This is referred to here as a plastic repulsion coefficient. For a very hard contact this plastic repulsion coefficient is infinitely small, i.e. \( \kappa_p \approx 0 \) and for a soft contact \( \kappa_p \rightarrow 1 \).

The Hamaker constant \( C_{H,\text{sls}} \) for solid-liquid-solid interaction (index sls) according to Lifshitz theory [211] is related to continuous media which depends on their permittivities (dielectric constants) and refractive indices [58]. The characteristic adhesion separation for a direct contact is of a molecular scale (atomic centre-to-centre distance) and can be estimated for the force equilibrium \( F = -dU/da = 0 \) of molecular attraction and repulsion potentials \( a = a_{F=0} \) [58, 178, 209].

This separation of the interaction potential minimum amounts to about \( a_{F=0} \approx 0.3 - 0.4 \) nm. With respect to particle surfaces this separation \( a_{F=0} \) depends on the properties of liquid-equivalent packed adsorbed water layers. Consequently, the particle contact behaviour is influenced by mobile adsorption layers due to condensed humidity of ambient air [240, 241, 247, 248, 253, 255].

This minimum centre separation \( a_{F=0} \) is assumed to be constant during loading and unloading, Fig. 8c), equivalent to stiff sphere repulsion of molecules [58].

If the contact circle radius \( r_K \) is small compared to the particle diameter \( d \), the elastic and plastic contact displacements can be combined and expressed with the annular elastic \( A_{el} \) and circular plastic \( A_{pl} \) contact area ratio according to Schubert et al. [143]:

\[ F_{H} = F_{H0} + \frac{p_{\text{vdW}}}{p_f \cdot (1 + 2 \cdot A_{pl}/(3 \cdot A_{el}))} \cdot F_N \]

For perfectly plastic contact displacement \( A_{el} \rightarrow 0 \) and one obtains again Eq. (72).

This linear increase of adhesion force \( F_H \) with increasing compressive normal force \( F_N \), Eqs. (71), (72) and (74), was experimentally confirmed for fine particles, e.g. by Schütz [240, 241] (\( \kappa_p = 0.3 \) for limestone) and Newton [242] (\( \kappa_p = 0.333 \) for poly(ethylene glycol), \( \kappa_p = 0.076 \) for starch, \( \kappa_p = 0.017 \) for lactose, \( \kappa_p = 0.016 \) for CaCO\(_3\)) with centrifuge tests [243] as well as by Singh et al. [245] (\( \kappa_p = 0.12 \) for poly(methylmethacrylate), \( \kappa_p \approx 0 \) for very hard sapphire, \( \alpha-\text{Al}_2\text{O}_3 \)) with an atomic force microscope (AFM). The two methods are compared with rigid and rough glass spheres (\( d = 0.1 \) to \( 10 \) µm), without any contact deformation, by Hoffmann et al. [217]. Additionally, using the isostatic tensile strength \( \sigma_0 \) determined by powder shear tests [98, 154, 179], this adhesion level is of the same order of magnitude as the average of centrifuge tests, see Spindler et al. [244].

### 3.3.3 Non-linear increase of adhesion force by normal force

The increase of adhesion force \( F_H \) due to contact compression was confirmed by Tabor [69], Maugis [224, 225] and Visser [246] as well. Also Maugis and Pollock [144] found that the separation was always brittle (index br) with a small initial slope of pull-off force, \( dF_{H,br}/dF_N \), for a comparatively small surface energy \( \sigma_{\text{ss}} \) of direct rigid sphere-gold plate contact. In contrast, a
pull-off force $F_{H,br}$ proportional to $\sqrt{F_N}$ was obtained from the Johnson’s contact theory [54] for the full plastic range of high loading and brittle separation of the contact [144], Table 1:

$$F_{H,br} = 2 \cdot \sigma_{ss} \cdot E^* \cdot \sqrt{\frac{F_N}{\pi \cdot p_f^3}}$$  \hspace{1cm} (75)

Mesarovic and Johnson [156] obtained an equivalent model for fully-plastic contact behaviour with elastic recovery, but with larger numerical factor $3 \pi/4$ compared to Eq. (75) (index MJ):

$$F_{H,MJ} = \frac{3}{2} \cdot \sigma_{ss} \cdot E^* \cdot \sqrt{\frac{\pi \cdot F_N}{p_f^3}}$$  \hspace{1cm} (76)

Substituting $F_N$ by normal force plus the contribution of short-range adhesion force outside the contact these Johnson-Maugis-Pollock and Mesarovic-Johnson models, Eqs. (75) and (76), were extended by Castellanos [161] (index JMPC):

$$F_{H,JMPC} = \frac{3}{2} \cdot \sigma_{ss} \cdot E^* \cdot \sqrt{\frac{\pi \cdot (F_N + 4 \cdot \pi \cdot r_{1,2} \cdot \sigma_{ss})}{p_f^3}}$$  \hspace{1cm} (77)

This model was experimentally confirmed by a fluidized bed test apparatus, with reverse gas permeation for initial pre-consolidation and following detachment and fluidization at very low stress levels [250, 251], for polymer particles by Castellanos et al. [252]. Recently, this load dependent adhesion force $F_H(F_N)$ was rewritten as [253]

$$F_H \propto F_N^n$$  \hspace{1cm} (78)

with an exponent $n = 0$ for the “classic” elastic contact of constant adhesion $F_{H0}$, $n = 1/3$ for predominant van der Waals interactions of the direct contact of spherical rough surfaces, i.e. plasticification of asperity tips [254], and $n = 1$ for moist contacts with liquid bridge bonds depending on the principal radii of meniscus curvature (bridge volume) relative to the roughness height.

Additionally, the load dependent adhesion force was also experimentally confirmed in wet environment of the particle contact by Butt et al. [214, 215, 216], Jones [256] and Higashitani et al. [257] with AFM measurements.

The dominant plastic contact deformation of surface asperities during the chemical-mechanical polishing process of silicon wafers was also observed, e.g., by Rimai and Busnaina [258], Ahmadi and Xia [259], Zhang and Busnaina et al. [261 - 263]. These particle-surface contacts and, consequently, asperity stressing by simultaneous normal pressure and shearing, contact deformation, microcrack initiation and propagation, and microfracture of brittle silicon asperity peaks affect directly the polishing performance. Thus the Coulomb friction becomes dominant also in wet environment.
3.4 Variation in adhesion due to inelastic contact flattening

3.4.1 Elastic-plastic force-displacement model for loading

All interparticle forces can be expressed in terms of a single potential function
\[ F_i = \pm \partial U_i(a_i) / \partial a_i, \quad (79) \]
e.g. the Mie potential [264], and thus are superposed. This is valid only for a conservative system in which the work done by the force \( F_i \) versus distance \( a_i \) is not dissipated as heat, but remains in the form of mechanical energy, simply in terms of irreversible deformation, e.g. initiation of nanoscale distortions, dislocations or lattice stacking faults. The overall potential function may be written as the sum of the potential energies of a single contact \( i \) and all particle pairs \( j \). Minimizing the potential function one obtains the potential force balance:
\[ \sum \sum_j \partial U_{ij} / \partial a_{ij} = 0 \quad (80) \]

This method was used to calculate the normal force - separation and displacement behaviours of an elastic contact of smooth spheres, see e.g. Derjaguin [110, 128], Bradley [112], Dahnecke [127], Maugis [59, 209] or Greenwood [147]. Continuum theory was the basis of JKR model [54, 126] which was extended to an elastic-plastic model by Mesarovic and Johnson [156] for unloading
\[ F_{N,\text{unload}} = \pi \cdot r_{K,f}^2 \cdot p_f \left[ \text{arcsin} \left( \frac{h_K}{h_{K,f}} \right) - \left( \frac{1 - h_K}{h_{K,f}} \right) \right] - 4 \pi \cdot \sigma_{ss} \cdot E^* \cdot \left( r_{1,2} \cdot h_K \right)^{1/2}, \quad (81) \]

which is rewritten here as normal force - displacement model using Eq. (6). But this model is limited to \( h_K \leq h_{K,f} \) which is not suitable to describe the contact behaviours of cohesive fine to ultrafine particles. The minimum of this function \( F_N(h_K) \) during unloading corresponds to the adhesion force Eq. (76) necessary to pull-off the particles each other or to break the contact.

On basis of numerical integration of the radial distribution of repulsive pressure \( p_{\text{rep}} \) by elastic and plastic stresses and attractive pressure \( p_{\text{at}} \) inside of the contact area
\[ F_N = 2 \pi \cdot \int_0^{h_K} p_{\text{rep}}(r_K^*) \cdot r_K^* \cdot dr_K^* + 2 \pi \cdot \int_0^{h_K} p_{\text{at}}(r_K^*) \cdot r_K^* \cdot dr_K^* \quad (82) \]
a comparatively sophisticated model for loading was recently published by Castellanos [161]. The kernels of the integrals are (\( r_K^* \) radial coordinate of contact area, \( r_{K,f} \) transition for yield according to Eq. (51), \( \Theta(\cdot) \) unit step function):
\[ p_{\text{rep}}(r_K^*) = p_m(r_K^*) \cdot \Theta(r_{K,f}^* - r_K^*) + \frac{E^*}{\pi \cdot r_{1,2}} \cdot \sqrt{r_K^* - r_{K,f}^*} \cdot \Theta(r_{K,f}^* - r_K^*) \quad (83) \]
Linear normal force-displacement model of load dependent adhesion

Neither the sophisticated functions of surface bending with tensile stresses at the contact perimeter [54, 109, 126, 144] nor the detailed calculations of pressure distribution \( p_m(r_K) \) within the core of the contact circle [54, 55, 156, 161], see Eqs. (54) and (84), have to be used in detail to calculate the load dependent adhesion by contact flattening of ultrafine cohesive particles with soft contacts. Taking into consideration the rule of minimum resistance in an inelastic cohesive powder, i.e. the normal stiffness is \( k_N \propto r_{1,2} \propto \text{particle size } d \), so that ultrafine particles, comparatively flexible embedded in a bulk or on surface layers, have better chances to respond softer or compliant than a fixed, relatively rigid indenter in any microhardness or AFM tests.

Thus, the simpler elastic-plastic force-displacement models for loading introduced by Schubert et al. [143], Eq. (74), and by Thornton [151], Eq. (86),

\[
p_{m}(r_K) = \begin{cases} 
0 & \text{elastic range} \\
\frac{E^*}{\pi \cdot r_{1,2}} \cdot \sqrt{r_K^2 - r_{K,f}^2} & \text{if } p_m < p_f = 2.8 \cdot \sigma_f \\
p_f & \text{otherwise}
\end{cases}
\]

and

\[
p_{H}(r_K^*) = \sqrt{\frac{2 \cdot \sigma_f \cdot E^* \cdot r_K}{\pi \cdot (r_K^2 - r_{K,f}^2)}}
\]

are the basis to extend their approaches by an extended attractive force contribution of contact flattening, see Tomas (1999/2001) [152 - 155]. The particle contact force equilibrium between attraction (-) and elastic plus, simultaneously, plastic repulsion (+) is calculated by \( r_K^* \) represents the coordinate of annular elastic contact area, i.e. for \( r_{K,f} \leq r_{K,pl} \leq r_K^* \leq r_K \):

\[
\sum F_{N,i} = 0 = -F_{H0} - p_{vdw} \cdot \pi \cdot r_K^2 - F_N + p_{pl} \cdot \pi \cdot r_{K,pl}^2 + 2 \cdot \pi \cdot \int_{r_{K,pl}}^{r_K} p_{pl}(r_K^*) \cdot r_K^* \, dr_K^*
\]

Superposition provided [205], the adhesion force \( -F_{H0} \) includes both the attraction at particle approach, i.e. short-range sphere-sphere interaction, and the contribution outside in the annular area which is located at the perimeter closed by the contact, Eq. (38). The term with the van der Waals pressure \( -p_{vdw} \cdot \pi \cdot r_K^2 \) models the effective attraction within the circular contact area between flattened smooth particle surfaces. The distribution of surface asperities, their local flattening and penetration is neglected at this model.

One has to solve the integral with the Hertzian contact pressure distribution Eq. (1):

\[
F_N + F_{H0} + p_{vdw} \cdot \pi \cdot r_K^2 = p_{pl} \cdot \pi \cdot r_{K,pl}^2 + 2 \cdot \pi \cdot p_{max} \cdot \int_{r_{K,pl}}^{r_K} r_K^* \cdot \sqrt{1 - \left( \frac{r_K^*}{r_K} \right)^2} \, dr_K^*
\]
And once more with Eq. (1) for the term in brackets one obtains:

\[ F_N + F_{h0} + p_{vdw} \cdot \pi \cdot r_k^2 = p_{pl} \cdot \pi \cdot r_{Kpl}^2 + \frac{2 \cdot \pi \cdot p_{max} \cdot r_k^2}{3} \left[ 1 - \left( \frac{r_{k,pl}}{r_k} \right)^2 \right]^{3/2} \]  

(89)

At the yield point \( r_{K,pl} = r_{K,f} \) the contact pressure reaches the plastic micro-yield strength in compression or microhardness \( p_{el} \leq p_{pl} \leq p_{f} = \text{const.} \) which is assumed to be constant.

\[ F_N + F_{h0} + p_{vdw} \cdot \pi \cdot r_k^2 = p_{pl} \cdot \pi \cdot r_{Kpl}^2 + \frac{2 \cdot \pi \cdot r_k^2 \cdot p_{el} \cdot \left( \frac{p_{el}}{p_{max}} \right)^2}{3} \]  

(90)

Because of plastic yielding without work-hardening effects for commonly non-metallic particles in powder processing, a pressure higher than \( p_{pl} \leq p_{f} \) is not possible (exceptionally Eq. (8) or see [156]) and thus, the fictitious contact pressure \( p_{max} \) is eliminated by Eq. (1):

\[ F_N + F_{h0} + p_{vdw} \cdot \pi \cdot r_k^2 = p_{f} \left[ \pi \cdot r_{Kpl}^2 + \frac{2 \cdot \pi \cdot r_k^2 \cdot p_{el}}{3 \cdot p_{pl}} \left( \frac{p_{el}}{p_{max}} \right)^2 \right] \]  

(91)

Finally, by the contact force equilibrium, Eq. (87),

\[ F_N + F_{h0} + p_{vdw} \cdot A_K = \pi \cdot p_{f} \cdot r_k^2 \left[ \frac{2}{3} + \frac{1}{3} \cdot \frac{r_{k,pl}^2}{r_k^2} \right] = p_{f} \cdot A_K \left[ \frac{2}{3} + \frac{1}{3} \cdot \frac{A_{pl}}{A_K} \right] \]  

(93)

the effective total contact area \( A_K \) is obtained:

\[ A_K = \frac{F_N + F_{h0}}{p_{f} \cdot \left[ \frac{2}{3} + \frac{1}{3} \cdot \frac{A_{pl}}{A_K} \right] - p_{vdw}} \]  

(94)

Next, the elastic-plastic contact area coefficient \( \kappa_A \) is introduced for the term in brackets. This dimensionless coefficient represents the ratio of plastic particle contact deformation area \( A_{pl} \) to total contact deformation area \( A_K = A_{pl} + A_{el} \) and includes a certain elastic displacement:

\[ \kappa_A = \frac{2}{3} + \frac{1}{3} \cdot \frac{A_{pl}}{A_K} \]  

(95)

The solely elastic contact deformation \( A_{el} = 0, \kappa_A = 2/3 \), has only minor relevance for cohesive powders in loading, Fig. 8, but for the complete plastic contact deformation \( A_{pl} = A_K \) the coefficient \( \kappa_A = 1 \) is obtained.

From Eqs. (73), (94) and (95) the sum of contact normal forces for loading is given as:

\[ F_N = \pi \cdot r_k^2 \cdot p_{f} \cdot (\kappa_A - \kappa_p) - F_{h0} \]  

(96)

From Eq. (5) the transition radius of elastic-plastic model \( r_{K,f,el-pl} \) (index el-pl) and from Eq. (6) the particle centre approach of the two particles \( h_{K,f,el-pl} \) are calculated as:
Checking this model, Eq. (97), with pure elastic contact deformation, i.e., $\kappa_p \rightarrow 0$ and $\kappa_A = 2/3$ the elastic transition radius $r_{K,f}$, Eq. (51), is also obtained.

For ultrafine limestone particles with the median diameter on mass basis (index 3) $d_{50,3} = 1.2 \, \mu m$ and effective constitutive parameters $E = 150 \, kN/mm^2$ modulus of elasticity, $\nu = 0.28$ Poisson ratio, $p_f = 300 \, N/mm^2$ micro-yield strength, $\kappa_A \approx 5/6$ contact area ratio, $\kappa_p = 0.153$ plastic repulsion coefficient) a contact radius of $r_{K,f,el-pl} = 1.8 \, nm$ and, from Eq. (98), a “homeopathic” centre approach of only $h_{K,f,el-pl} = 0.011 \, nm$ are obtained. That is a very small indentation for the yield point $Y$, Fig. 8, calculated, in principle, by means of a continuum approach of molecular surface properties. This $h_{K,f,el-pl}$ is a bit shifted to the left compared with the displacement limit $h_{K,f}$ according to Eq. (52). The contact displacement for yield is equivalent to an effective microscopic normal force of $F_{N,f,el-pl} = - 0.62 \, nN$ or to a small macroscopic stress level of about $\sigma = - 0.15 \, kPa$ (with porosity $\varepsilon_0 = 0.74$) in powder storage, handling and processing.

Introducing the particle centre approach of the two particles Eq. (6) in Eq. (96), a very useful linear force-displacement model is obtained for contact loading at $\kappa_A \approx const$:

$$ F_N = \pi \cdot r_{1,2} \cdot p_f \cdot (\kappa_A - \kappa_p) \cdot h_k - F_{H0}, \quad (99) $$

This straight-line model of elastic-plastic yield limit is shown in Fig. 8 for limestone powder which was back-calculated from material data and shear test data [154, 271].

The previous contact model can be supplemented by viscoplastic stress-strain behaviour, i.e. strain rate dependence on initial yield stress. One obtains for elastic-viscoplastic contact deformation with Eqs. (60) and (99) ($\kappa_A \approx const.$):

$$ F_N = \pi \cdot r_{1,2} \cdot \eta_k \cdot (\kappa_{A,1} - \kappa_{p,t}) \cdot h_k - F_{H0} \quad (100) $$

A dimensionless viscoplastic contact repulsion coefficient $\kappa_{p,t}$ is introduced as the ratio of the van der Waals attraction to viscoplastic repulsion effects which are additionally acting in the contact after attaining the maximum pressure for yielding.

$$ \kappa_{p,t} = \frac{p_{vdw}}{\eta_k \cdot h_k} \quad (101) $$

The consequences for the variation in adhesion force are discussed in section 3.4.3 [154].

### 3.4.2 Unloading and reloading hysteresis and contact detachment

Constant mechanical properties provided, the finer the particles the smaller is the yield point $Y$ in Fig. 7, see Eq. (52) [179]. An initial and exclusive elastic contact deformation at loading has no relevance for the irreversible consolidation of fine to ultrafine powders and can be excluded.
here. One has to consider an elastic-plastic contact behaviour which is characterised by the elastic-plastic yield limit, Eq. (99). This line cannot be crossed. Below the yield limit the elastic domain begins. The model is equivalent to the yield surface in continuum mechanics. Any load $F_N$ yields an increasing displacement $h_K$.

But, if one would start to unload, beginning at arbitrary point $U$, the elastically deformed, annular contact zone recovers along a curve $U - A$, see Fig. 9, Tomas (2000/2001) [154, 155].

Fig. 9: Representative particle contact deformation – elastic unloading and reloading with dissipation (limestone, $d_{50,3} = 1.2 \mu m$) [155]. Below the elastic-plastic yield limit the elastic domain begins. After unloading $U - E$ the contact recovers elastically in the compression mode and remains with a plastic displacement $h_{K,E}$. Below point $E$ on the axis the tension mode begins. Between the points $U - E - A$ the contact recovers elastically according to Eq. (104) to a displacement $h_{K,A}$. The reloading curve runs from point $A$ to $U$ to the displacement $h_{K,U}$, Eq. (109).

If one assumes a comparatively small total contact area, this unload function is designed to be an extended Hertz law, see Thornton [151]. This contact area has changed the radius of effective curvature $r_{1,2}$ at incipient loading to a larger radius of unloaded curvature $r_{1,2,U}$ by the irreversible flattening of the core zone. Provided that during elastic unloading, the changes in contact region are geometrically similar to the changes that occur during loading $h_{K,U}/r_{1,2,U} = h_{K,f}/r_{1,2}$ the following ratio was derived by Stronge [55]:

$$\frac{r_{1,2,U}}{r_{1,2}} = \frac{2 \cdot h_{K,U}}{h_{K,f}} - 1$$  (102)

Instead of $r_{1,2}$ this new curvature radius $r_{1,2,U}$ can be used in Eqs. (104) and (109) to adjust the unloading/reloading functions stiffer than shown in Fig. 9.
At a series arrangement of elastic and plastic deformation elements the total displacement $h_{K,U}$ is the sum of elastic $h_{K,el}$ and plastic displacements $h_{K,A}$. Thus using this balance, the elastic displacement is replaced in Eq. (7) by ($h_K$ is written here without index U):

$$h_{K,el} = h_K - h_{K,A}$$ (103)

For a parallel arrangement of repulsive elastic and attractive bond elements (molecules) in the stressed and strained contact, the total force $F_{N,unload}$ is superimposed by an elastic $F_{N,el}$ and a minimum normal (tensile) force or “adhesion limit” $F_{N,Z}(h_{K,A})$ contribution. This adhesion limit, shown as blue line in Fig. 10, can not be crossed as well. Thus, the normal force balance yields:

$$F_{N,unload} = \frac{2}{3} E^* \sqrt{f_{1,2} \cdot (h_K - h_{K,A})^3} + F_{N,Z}(h_{K,A})$$ (104)

Fig. 10: Representative particle contact deformation – a) to d) the complete survey of loading, unloading and detachment behaviours of limestone [155]. Pressure and compression are defined as positive but tension and extension are negative, above panel. After approaching from an infinite distance $\rightarrow \infty$ to this minimum centre separation $a_{F=0}$ the sphere-sphere contact is formed without any contact deformation by the attractive short-range adhesion force $F_{H0}$ (the so-called “jump in”). As the response, from $F_{H0} - Y$ the contact is elastically compacted, forms an approximated circular contact area, panel b) and starts at the yield point Y at $p_{max} = p_f$ with plastic yielding, panel c). This flattening is expressed by annular elastic $A_{el}$ and circular plastic $A_{pl}$ contact area. Between the elastic-plastic yield limit and the adhesion limit the elastic domain is located. If the contact is unloaded the contact recovers elastically according to Eq. (104) to a displacement $h_{K,A}$. If one applies a certain pull-off force $F_{N,Z}$ the adhesion limit at failure point A is reached, Eq. (112), and the contact plates fail and detach with the increasing distance $a = a_{F=0} + h_{K,A} - h_K$ as given in Eq. (114), panel d). The effective material data are: modulus of elasticity $E = 150$ kN/mm$^2$, plastic micro-yield strength $p_f = 350$ N/mm$^2$, Poisson ratio $\nu = 0.28$, Hamaker constant $C_{H,sls} = 3.8 \times 10^{-20}$ J, equilibrium centre separation for dipole interaction $a_{F=0} = 0.336$ nm, elastic-plastic contact area coefficient $\kappa_A = 5/6$ and the plastic repulsion coefficient $\kappa_p = 0.153$. 
Between the points U – E, Fig. 9, the contact recovers elastically along this extended Hertzian curve, Eq. (104), down to the perfect plastic displacement, \( h_{K,E} \), obtained with Eq. (99):

\[
h_{K,E} = h_{K,U} - \frac{2}{3} h_{K,U} \cdot \left( h_{K,U} - h_{K,U}^2 \right) \tag{105}\]

Thus, the contact area ratio \( \kappa_A \) is expressed more in detail with Eqs. (6) and (95) for elastic \( \kappa_A = 2/3 \) and perfect plastic contact deformation, \( \kappa_A = 1 \) if \( h_{K,U} \rightarrow \infty \):

\[
\kappa_A = 2 + \frac{h_{K,E}}{3 \cdot h_{K,U}} = 1 - \frac{1}{3} \sqrt{\frac{h_{K,f}}{h_{K,U}}} \tag{106}\]

If this contact area ratio of Eq. (106) is used in the elastic-plastic yield limit Eq. (99), a slightly nonlinear (progressively increasing) curve for loading is obtained as well:

\[
F_N = \pi \cdot r_{i,2} \cdot p_f \cdot \left( 1 - \frac{1}{3} \sqrt{\frac{h_{K,f}}{h_K}} - \kappa_p \right) \cdot h_k - F_{H0} \tag{107}\]

The contact stiffness of the non-linear elastic-plastic yield limit is for \( \kappa_A = f(h_K) \) with Eq. (106):

\[
k_{N,el-pl} = \pi \cdot r_{i,2} \cdot p_f \cdot \left( 1 - \frac{2}{9} \sqrt{\frac{h_{K,f}}{h_K}} - \kappa_p \right) \approx \pi \cdot r_{i,2} \cdot p_f \cdot (\kappa_A - \kappa_p) \tag{108}\]

The right term is valid for the linear elastic-plastic yield limit (\( \kappa_A = \text{const.} \), Eq. (99).

At the transition point \( h_K = h_{K,A} \) of the unload curve, Eq. (104), the plate-plate contact remains just adhered with a “frozen” radius \( r_{K,A} \) or plastic displacement \( h_{K,A} \) and zero unload stiffness. The reloading would run along an equivalent curve

\[
F_{N,\text{reload}} = -\frac{2}{3} \cdot E_0 \cdot \sqrt{r_{i,2} \cdot \left( h_{K,U} - h_k \right)^3} + F_{N,U}(h_{K,U}) \tag{109}\]

from point A to point U forward to the displacement \( h_{K,U} \) as well, Fig. 9. The displacement \( h_{K,A} \) at point A of incipient contact detachment is calculated from Eqs. (98), (99), (104) and (112) as an implied function (index (0) for beginning iterations) of the displacement history point \( h_{K,U} \):

\[
h_{K,A,(0)} = h_{K,U} - \frac{1}{3} \sqrt{h_{K,U} \cdot \left( h_{K,A} + \kappa \cdot h_{K,A,(0)} \right)^2} \tag{110}\]

If one replaces the contact radius \( r_{K,A}^2 = r_{i,2} \cdot h_{K,A} \) after contact flattening in the sum of attractive particle contact forces, as given from Eq. (87),

\[
\sum F_{at} = 0 = -F_{H0} - p_{Vdw} \cdot \pi \cdot r_{K,A}^2 - F_{N,Z}(h_{K,A}) \tag{111}\]

the force-displacement relation \( F_{N,Z}(h_{K,A}) \) for attraction or adhesion by molecular interactions is obtained, the so-called adhesion limit (here negative), blue line in Fig. 10:

\[
F_{N,Z}(h_{K,A}) = -\pi \cdot r_{i,2} \cdot p_{Vdw} \cdot h_{K,A} - F_{H0} \tag{112}\]

The adhesion stiffness of this maximum pull-off force results in:

\[
k_{N,A} = -\pi \cdot r_{i,2} \cdot p_{Vdw} \tag{113}\]
For plausibility, the absolute value of this adhesion stiffness \( k_{N,A} \) is similar to the contact stiffness \( k_{N,pl} \) of perfect plastic repulsion behaviour, Eq. (58). It is worth to note here, that the elastic-plastic repulsion or yield limit, black line in Fig. 10, and the adhesion limit by van der Waals attraction are non-symmetric with different slopes.

When the adhesion limit or incipient contact failure at point A in the diagram of Fig. 10 is reached the contact plates detach with the increasing distance \( a = a_{F=0} + h_{K,A} - h_K \) [181]. This actual short-range particle separation can be calculated by the sum of a hyperbolic adhesion force curve \( p_{VdW}(a) \propto a^{-3} \) from the plate-plate model as given from Eq. (73) and \( F_{H0}(a) \propto a^{-2} \) according to the sphere-sphere model of adhesion force from Eq. (158):

\[
F_{N,Z}(h_K) = -\frac{F_{H0}}{(1 - \frac{h_K}{a_{F=0}})^2} - \frac{\pi \cdot r_{1,2} \cdot p_{VdW} \cdot h_{K,A}}{\left(1 + \frac{h_{K,A}}{a_{F=0}} - \frac{h_K}{a_{F=0}}\right)^3} \tag{114}
\]

This is the correct model Eq. (114) for short-range contact detachment or decohesion, see orange curve in Fig. 10, which replaces the incorrect Eq. (68) in [181] and Eq. (17) in [185] as given in these previous papers.

The complete survey of loading, unloading, reloading, dissipation and detachment behaviours of limestone is shown in Fig. 10 as a combination of Fig. 7a) to Fig. 9d). For a better survey, the necessary equations are compiled in Table 2.

Table 2: Compilation of normal force – displacement models \( F_N(h_K) \) of load dependent van der Waals adhesion for particle approach, loading, elastic-plastic contact deformation (flattening), unloading, reloading and contact detachment, see Fig. 10.

<table>
<thead>
<tr>
<th>Process</th>
<th>Model</th>
<th>Equation</th>
<th>Eq.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Approach - ( \infty &lt; h_K \leq 0 )</td>
<td>sphere-sphere</td>
<td>( F_{H0} = \frac{C_{H,slr} \cdot h_{1,2}}{a_{F=0} - h_K} \left[ 1 + \frac{r_{1,2}/h_{1,2}}{(1 + 2 \cdot r_{1,2}/(a_{F=0} - h_K))^2} \right] )</td>
<td>(158)</td>
</tr>
<tr>
<td>Elastic deform. ( 0 \leq h_K \leq h_{K,f} )</td>
<td>Hertz [108]</td>
<td>( F_N = \frac{2}{3} \cdot E^* \cdot \sqrt{r_{1,2} \cdot h_K^3} )</td>
<td>(7)</td>
</tr>
<tr>
<td>Yield limit ( h_{K,f} \leq h_K \leq h_{K,U} )</td>
<td>elastic-plastic</td>
<td>( F_N = \pi \cdot r_{1,2} \cdot p_f \cdot (\kappa_A - \kappa_p) \cdot h_K - F_{H0} )</td>
<td>(99)</td>
</tr>
<tr>
<td>Unload ( h_{K,A} \leq h_K \leq h_{K,U} )</td>
<td>elastic recovery detachment</td>
<td>( h_{K,A,(i)} = h_{K,U} - \frac{3}{\sqrt{h_{K,U}^2 + \kappa \cdot h_{K,A,(0)}^2}} )</td>
<td>(104)</td>
</tr>
<tr>
<td>Reload ( h_{K,A} \leq h_K \leq h_{K,U} )</td>
<td>elastic, dissipative</td>
<td>( F_N = \frac{-2}{3} \cdot E^* \cdot \sqrt{r_{1,2} (h_{K,U} - h_K)^3} + \pi \cdot r_{1,2} \cdot p_f (\kappa_A - \kappa_p) \cdot h_{K,U} - F_{H0} )</td>
<td>(109)</td>
</tr>
<tr>
<td>Adhesion limit ( 0 \leq h_K \leq h_{K,A} )</td>
<td>plate-plate</td>
<td>( F_N = -\pi \cdot r_{1,2} \cdot p_{VdW} \cdot h_K - F_{H0} )</td>
<td>(112)</td>
</tr>
<tr>
<td>Detachment - ( \infty &lt; h_K \leq h_{K,A} )</td>
<td>plate-plate</td>
<td>( F_N(h_K) = -\frac{F_{H0} \cdot a_{F=0}^2}{(a_{F=0} - h_K)^2} - \frac{\pi \cdot r_{1,2} \cdot p_{VdW} \cdot h_{K,A}}{(a_{F=0} + h_{K,A} - h_K)^3} \cdot a_{F=0}^3 )</td>
<td>(114)</td>
</tr>
</tbody>
</table>
Using Eqs. (99), (110) and (112), one may calculate a secant unload stiffness $k_{N,U}$ of the elastic domain between the elastic-plastic yield limit point U and the adhesion limit point A, first used by Luding [157, 158, 194]:

$$k_{N,U} = \frac{F_{N,U} - F_{N,A}}{h_{K,U} - h_{K,A}} = \pi \cdot r_{1,2} \cdot p_f \cdot \left[ \frac{\kappa_A \cdot h_{K,U}}{h_{K,U} - h_{K,A}} - \kappa_f \right]$$

(115)

Using this slope Eq. (115) and Eq. (99) for the given point U a simple linear force – displacement function for unload, besides the nonlinear Eq. (104), is calculated:

$$F_{N,unload} = \pi \cdot r_{1,2} \cdot p_f \cdot \left[ \frac{\kappa_A \cdot h_{K,U}}{h_{K,U} - h_{K,A}} \cdot \left[ h_K - h_{K,A}\right] - \kappa_f \cdot h_K \right] - F_{H0}$$

(116)

It is worth to note here that this averaged unload stiffness is increasing with increasing contact flattening $h_{K,U}$, see the steeper unload curves in Fig. 11 with increasing load $F_N$.

These theoretical predictions of normal force-displacement behaviours are very useful to describe the compression of primary elastic, elastic-plastic particle compounds or elastic-plastic granules at conveying and handling operations. Elastic-plastic load and Hertzian unload curves, Eq. (99) and Eq. (104), and the unload/reload hysteresis were experimentally confirmed, e.g. for coarse granules [265, 266, 267]. Moreover, Horn et al. [268] measured an inelastic load/unload hysteresis between two mica sheets by Israelachvili’s surface force apparatus [280].

Fig. 11: Force - displacement diagram of back-calculated representative contact deformation of cohesive limestone particles as spheres, median diameter $d_{50} = 1.2 \mu m$. This hysteretic behaviour could be shifted along the elastic-plastic limit and depends on the pre-loading, or in other words, preconsolidation level. Thus, the variation in adhesion forces between particles depends directly on this frozen irreversible deformation, the so-called contact preconsolidation history.
3.4.3 Viscoplastic contact behaviour and time dependent consolidation

An elastic-plastic contact may be additionally deformed during the indentation time, e.g., by viscoplastic flow, section 3.2.5. Thus, the adhesion force increases with interaction time [75, 57, 281, 315]. This time dependent consolidation behaviour (index t) of particle contacts in a powder bulk was previously described by a parallel series (summation) of adhesion forces, see Table 1, last line marked with Tomas [98, 152, 269, 270, 271]. This previous method refers more to incipient sintering or contact fusion of a thermally sensitive particle material [166] without interstitial adsorption layers. This micro-process is very temperature sensitive [98, 152, 269, 270].

Additionally, the increasing adhesion may be considered in terms of a sequence of rheological models as the sum of resistances due to plastic and viscoplastic repulsion \( \kappa_p + \kappa_{p,t} \), 4th line in Table 3. Hence the repulsion effect of “cold” viscous flow of comparatively strongly bonded adsorption layers on the particle surface is taken into consideration (the limestone powder has a specific surface area of \( A_{S,m} = 9.2 \text{ m}^2/\text{g} \) with a certain water adsorption capacity). This rheological model is only valid for a short term indentation of \( \eta_k \cdot \tau \), e.g. \( \tau < 61 \text{ h} \) for the cohesive limestone powder. All the material parameters are collected in Table 3.

Table 3: Material parameters of load dependent adhesion force functions \( F_{H}(F_N) \) in Fig. 17 and contact friction

<table>
<thead>
<tr>
<th>.instantaneous contact consolidation</th>
<th>Eq.</th>
<th>time dependent viscous contact consolidation</th>
<th>Eq.</th>
</tr>
</thead>
<tbody>
<tr>
<td>plastic repulsion coefficient</td>
<td>( \kappa_p = \frac{p_{\text{VdW}}}{p_f} = \frac{C_{\text{H,sls}}}{6 \cdot \pi \cdot a_{\text{p,0}} \cdot p_f} )</td>
<td>(73)</td>
<td>( \kappa_{p,t} = \frac{p_{\text{VdW}}}{\eta_k} \cdot t )</td>
</tr>
<tr>
<td>elastic-plastic contact area coefficient</td>
<td>( \kappa_A = \frac{2}{3} + \frac{A_{\text{pl}}}{3(A_{\text{pl}} + A_{\text{el}})} )</td>
<td>(95)</td>
<td>( \kappa_{A,t} = \frac{2}{3} + \frac{A_{\text{pl}} + A_{\text{vis}}}{3(A_{\text{pl}} + A_{\text{vis}} + A_{\text{el}})} )</td>
</tr>
<tr>
<td>elastic-plastic contact consolidation coefficient</td>
<td>( \kappa = \frac{\kappa_p}{\kappa_A - \kappa_p} )</td>
<td>(151)</td>
<td>( \kappa_{\text{vis}} = \frac{\kappa_p + \kappa_{p,t}}{\kappa_{A,t} - \kappa_p - \kappa_{p,t}} )</td>
</tr>
<tr>
<td>averaged particle radius</td>
<td>( r_{1,2} = (1/r_1 + 1/r_2)^{-1} )</td>
<td>(3)</td>
<td>-</td>
</tr>
<tr>
<td>averaged modulus of elasticity</td>
<td>( E^* = 2 \cdot \left( \frac{1 - v^2_i}{E_1} + \frac{1 - v^2_j}{E_2} \right)^{-1} )</td>
<td>(4)</td>
<td>-</td>
</tr>
<tr>
<td>Poisson ratio</td>
<td>( \nu )</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>micro-yield strength of contact</td>
<td>( p_f )</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>apparent contact viscosity</td>
<td>-</td>
<td>-</td>
<td>( \eta_k )</td>
</tr>
<tr>
<td>Adhesion force of sphere-sphere contact</td>
<td>( F_{H0} )</td>
<td>(158)</td>
<td>-</td>
</tr>
<tr>
<td>contact friction coefficient</td>
<td>( \frac{\sum F_S}{\sum F_N} = \tan \varphi_i )</td>
<td>(211)</td>
<td>( \frac{214}{\sum F_N} )</td>
</tr>
</tbody>
</table>
A viscoelastic relaxation in the particle contact may be added as a time dependent function of the average modulus of elasticity $E^*$, Yang [165] and Krupp [57] ($t_{\text{relax}}$ is the characteristic relaxation time):

$$\frac{1}{E^*} = \frac{1}{E^*_{\infty}(t \to \infty)} + \left( \frac{1}{E^*_0(t = 0)} - \frac{1}{E^*_{\infty}} \right) \cdot \exp\left(-\frac{t}{t_{\text{relax}}}\right)$$

(117)

The slopes of the elastic-plastic, viscoelastic-viscoplastic yield and adhesion limits as well as the unloading and reloading curves, which include a certain relaxation effect, are influenced by the increasing softness or compliance of the contact between spherical particles with loading time, Fig. 12. This model system includes all the essential constitutive functions of the authors named before [108, 133, 143, 151, 165]. A survey of the essential contact force-displacement models is given in Fig. 5 and Table 1.

Obviously, contact stiffness, deformation, contact and adhesion forces are stochastically distributed material functions. Usually one may focus here only on the characteristic or averaged values of these constitutive functions.

![Diagram](https://example.com/diagram.png)

Fig. 12: Representative elastic-plastic, viscoelastic-viscoplastic particle contact deformations – (limestone, median particle size $d_{50,3} = 1.2 \, \mu m$, surface diameter $d_s = 240 \, nm$, specific surface area $A_{S,m} = 9.2 \, m^2/g$, solid density $\rho_s = 2740 \, kg/m^3$, surface moisture $X_W = 0.5 \, %$, temperature $\theta = 20^\circ C$, loading time $t = 24 \, h$). The material data, modulus of elasticity $E = 150 \, kN/mm^2$, modulus of relaxation $E_\infty = 130 \, kN/mm^2$, relaxation time $t_{\text{relax}} = 2.4 \, \text{min}$, plastic micro-yield strength $p_f = 300 \, N/mm^2$, contact viscosity $\eta_K = 1.7 \times 10^{13} \, \text{Pa}s$, Poisson ratio $\nu = 0.28$, Hamaker constant $C_{H,sls} = 3.8 \times 10^{-20} \, J$, equilibrium centre separation for dipole interaction $d_F = 0 = 0.336 \, nm$, contact area ratio $\kappa_A = 5/6$ are assumed as appropriate for the characteristic contact properties. The plastic repulsion coefficient $\kappa_p = 0.153$ and viscoplastic repulsion coefficient $\kappa_{p,t} = 0.27$ are back-calculated from shear test data in a powder continuum [154, 271].
3.5 Measurement of adhesion forces

3.5.1 Adhesion effects between particles and surfaces

There are a lot of physical effects which are directly related to adhesion, e.g., elasticity, plastic yield strength, viscosity or phase conversion enthalpies, see e.g. [5, 58, 178]. Especially, the collective molecular interactions tend to be of a thermodynamic nature and are not directly translatable into a force-displacement/separation model which we are interested in this paper, see the examples described by Israelachvili [58].

Particle detachment and peeling experiments, Fig. 13a, b) provide information on particle adhesion forces and the adhesion energies of solid surfaces in contact (i.e., attractive short-range forces). Such experiments are important in powder technology, xerography, ceramic processing, the making of adhesive films and in understanding how cracks propagate in solids.

Fig. 13: Adhesion effects between particles and surfaces by intermolecular forces according to Israelachvili [58]. Different types of effects and testing techniques that provide information on the forces between particles and surfaces:

a) Adhesion measurements (practical applications: xerography, particle adhesion, powder technology, ceramic processing).

b) Peeling measurements (practical applications: adhesive tapes, materials fracture and crack propagation).

c) Direct measurements of force as a function of surface separation (practical applications: testing theories of intermolecular forces).

d) Contact angle measurements (practical applications: testing wettability and stability of surface films, detergency).
c) Equilibrium thickness of thin free film (practical applications: soap films, foams).

d) Equilibrium thickness of thin adsorbed films (practical applications: wetting of hydrophilic surfaces by water, adsorption of molecules from vapour, protective surface coatings and lubricant layers, photographic films).

e) Interparticle spacing in liquids (practical applications: colloidal suspensions, paints, pharmaceutical dispersions).

f) Sheet-like particle spacing in liquids (practical applications: clay and soil swelling behaviour, microstructure of soaps and biological membranes).

g) Coagulation studies (practical application: basic experimental technique for testing the stability of colloidal preparations).

Measuring the force between two macroscopic surfaces as a function of surface separation can provide the full force law of an interaction, Fig. 13c). Such direct force measurements are described in the next section 3.5.2.

Various surface studies such as surface tension and contact angle measurements give information on liquid-liquid and solid-liquid adhesion energies, Fig. 13d). When contact angles are measured under different atmospheric environments or as a function of time, these relatively simple experiments can provide surprisingly valuable insights into the states of surfaces and adsorbed films, and of molecular reorientation times at interfaces.

The thickness of free soap films and liquid films adsorbed on surfaces Fig. 13e, f) can be measured as a function of salt concentration or vapour pressure. Such experiments provide information on the long-range repulsive forces stabilizing thick wetting films. Various optical techniques (e.g., reflected intensity, total internal reflection spectroscopy) have been used to measure film thickness to within 0.1 nm.

Dynamic interparticle separations and motions in liquids can be measured using NMR, light scattering, x-ray scattering and neutron scattering Fig. 13g, h). In such experiments the particles can be globular or spherical (e.g., clays, lipid bilayers), or rod-like (e.g., DNA). The interparticle forces can be varied by changing the solution conditions, and their mean separation can be varied by changing the quantity of solvent, for example, by changing the hydrostatic or osmotic pressure via a semipermeable membrane.

In coagulation studies on colloidal dispersions Fig. 13i) the salt concentration, pH, or temperature of the suspending liquid medium (usually water) is changed until the dispersion becomes unstable and the particles coalesce (coagulate or flocculate). Coagulation rates can be very fast or very slow. Such studies provide information on the interplay of repulsive and attractive forces between particles in pure liquids as well as in surfactant and polymer solutions.

3.5.2 Principles of adhesion force testing methods

Fig. 14 shows the principles of measuring methods of adhesion force, which are based in applying external force on the particles. The interacting force between two microscopic bodies is measured by the spring balance method as a function of the elongation of the spring when they
are separated. First of all, this method was used by Bradley [111], then by Derjaguin et al. [273], Overbeek et al. [274, 275], Israelachvili and Tabor [278, 279], Israelachvili and Adams [280] and Israelachvili with his surface force apparatus (SFA) [58].

In the centrifugal method, the plate on which particles are deposited is put on a centrifuge to press particles on the surface by a compressive force $F_C$, and after this so-called contact preconsolidation, to detach the particles [57]. The centrifugal force where half of the deposited particles are removed is measured to evaluate the average adhesion force [239 - 241]. When one turns the plate perpendicular to the axis of revolutions, the tangential or shear force distribution of the contacts can be measured by the cumulative mass fraction of detaching particles.

![Diagram of adhesion force measurement methods](Mechanics_Particle_Adhesion_full.doc)

**Fig. 14:** Testing the adhesion force between particle and surface collected by Masuda [105].

The vibration method, first described by Derjaguin [277], is based on particle detachment from a vibrating surface caused by its inertia at a certain acceleration. Thus, the vibration does not only yield a detachment or pull-off force to compensate the adhesion force, but also causes compressive normal forces between particles and surface of the same order. For example, this alternating contact compression and detachment forces are frequently used during the dynamic stressing of cohesive powders as flow promotion in the practice of process engineering [330].

The impact separation method uses an acceleration generated by the bullet [276] or hammer impact [282]. When a fluid flow field is applied to particles adhered on a plate, the particles suffer a force caused by the flow. At a certain flow velocity, particles start to detach from the plate. Then, the adhesion force can be obtained as a function of flow velocity and/or stress [283].

Vibration and hydrodynamic method were recently combined. Particle detachment events are continuously recorded and correlated with acting acceleration, particle mass and flow conditions, which allows to calculating the pull-off force [284].

In a direct force-separation measurement a so-called atomic force microscope (AFM) is used [285, 286]. The particle is stuck at a cantilever, the so-called “colloid probe technique” [214],
Fig. 15 centre panel. The sample is moved up and down against the fixed cantilever by applying a voltage to the piezoelectric translator. The cantilever deflection $\Delta a_c$ is measured versus position of the piezo $\Delta a_p$ normal to the surface. To obtain a force distance curve $\Delta a_c$ and $\Delta a_p$ have to be converted into normal force and separation (distance). The normal force $F_N = k_c \cdot \Delta a_c$ is obtained by multiplying the deflection of the cantilever with its spring constant $k_c$, and the tip–sample separation $D = \Delta a_p + \Delta a_c$ is calculated by adding the deflection $\Delta a_c$ to the position $\Delta a_p$.

Fig. 15: Measurements of adhesion force - separation function by Particle Interaction Apparatus (PIA) according to Butt et al. [214]. The primary result is a plot of the deflection of the cantilever $\Delta a_c$ versus the height position of the sample, $\Delta a_p$ (right). A cycle in the force measurement starts at a large tip-surface separation. At large distances no force acts between tip and sample; the cantilever is not deflected. In the scheme it was assumed that at smaller distances a repulsive force acts between tip and sample. Hence, when the sample approaches the tip the cantilever bends upwards. Since tip and sample are not in contact this region is often referred to as the non-contact region. At a certain point the tip often jumps onto the sample surface. This “jump-in” occurs when the gradient of attractive force exceeds the spring constant plus the gradient of repulsive forces. Moving the sample still further causes a deflection of the cantilever of the same amount as the sample is moved. This “contact region” which includes contact loading is represented by the vertical line. Finally the sample is withdrawn at point U and moves to the detachment point A. Finally its starting position is reached. During retraction the tip often sticks to the surface up to large distances due to adhesion. To obtain force-separation curves the original deflection-position curves have to be converted [214].

The deflection of the cantilever is normally measured using the optical lever technique. A beam from a laser diode is focused onto the end of the cantilever and the position of the reflected beam is monitored by a position sensitive detector array (photodetector). The backside of the cantilever is usually covered with a thin gold layer to enhance its reflectivity. When a force is applied to the probe, the cantilever bends and the reflected light beam moves through a certain angle.
The resolution of the optical lever technique is roughly $10^{-13} \cdot \Delta w / \sqrt{\Delta t}$ ($\Delta w$ pixel width at photosensor, $\Delta t$ time for measuring a pixel of the force curve). With typically $\Delta t = 0.1$ ms the height-position resolution is 0.01 nm. But in practice, the position sensitivity is often limited by thermal cantilever vibrations, which are $\sqrt{4 \cdot k_B T / (3 \cdot k_c)}$ ($k_B$ Boltzmann constant, $T$ temperature and $k_c$ spring constant of the cantilever). With typical spring constants between 0.01 – 1 N/m the amplitude of thermal noise is 0.7 - 0.07 nm at room temperature.

Cantilevers for AFMs are usually V-shaped to increase their lateral stiffness. They are typically 100 - 200 $\mu$m long, each arm is about 20 $\mu$m wide and $h = 0.5$ $\mu$m thick. The spring constant of V-shaped cantilevers is often approximated by that of a rectangular bar of twice the width of each arm, for details see Butt et al. [214].

Friction forces as function of normal forces can also be measured by AFM equipment, e.g. [212, 213, 218, 256].

### 3.6 Energy absorption in a contact with adhesion and particle impact

#### 3.6.1 Dissipative behaviour of contact unload/reload

If one assumes a single elastic-plastic particle contact as a conservative mechanical system without heat dissipation, the energy absorption equals one closed cycle of the lens-shaped area between the unloading and reloading curves from point U to A as shown in Fig. 9:

$$W_{\text{diss}} = \frac{h_{K_U}}{C} \int_{h_{K_A}}^{h_{K_U}} F_N(h_K) \, dh_K = \int_{h_{K_A}}^{h_{K_U}} F_{N,\text{load}}(h_K) \, dh_K - \int_{h_{K_A}}^{h_{K_U}} F_{N,\text{unload}}(h_K) \, dh_K$$

(118)

from Eqs. (104), (112) for $F_{H,A}$ and (109), (99) for $F_{N,U}$ one obtains finally:

$$W_{\text{diss}} = -\frac{8 \cdot E^*}{15} \sqrt{r_{1,2} \cdot (h_{K,U} - h_{K,A})^3} + \pi \cdot r_{1,2} \cdot \rho_f \cdot \left[ \kappa_A \cdot h_{K,U} - \kappa_p \cdot (h_{K,U} - h_{K,A}) \right] \cdot (h_{K,U} - h_{K,A})$$

(119)

Additionally, the specific or mass related energy absorption $W_{m,\text{diss}} = k \cdot W_{\text{diss}} / m_{1,2}$ is often used to evaluate this contribution of energy consumption in a bulk powder by a comfortable micro-macro transition. The average mass of contacting particles 1 and 2 is similar to Eq.(3) [4]:

$$m_{1,2} = \frac{m_1 \cdot m_2}{m_1 + m_2} = \left( \frac{1}{m_1} + \frac{1}{m_2} \right)^{-1}$$

(120)

But to demonstrate directly the influence of the main particle property, i.e. the averaged particle size $d = 4r_{1,2}$, this average particle mass $m_{1,2}$ is approximated as ($\rho_s = \text{const}$):

$$m_{1,2} \approx \frac{8 \cdot \pi}{3} \cdot \rho_s \cdot r_{1,2}^3$$

(121)

Additionally, an averaged or characteristic contact number in the bulk powder (coordination number $k \approx \pi/e$ [18]) is used and one obtains finally:
\[ W_{\text{m,diss}} = -E^* \left( \frac{h_{K,U} - h_{K,A}}{r_{1,2}} \right)^{5/2} + \frac{3 \cdot \pi \cdot p_f \left( h_{K,U} - h_{K,A} \right)}{8 \cdot r_{1,2}^2 \cdot \varepsilon \cdot \rho_s} \left[ k_A \cdot h_{K,U} - k_p \cdot \left( h_{K,U} - h_{K,A} \right) \right] \] (122)

For example, a specific energy absorption of 2 to 41 µJ/g was dissipated during a single unloading - reloading cycle in the limestone bulk powder with average pressures of only \( \sigma_{M,st} = 3 \) to 25 kPa (or major principal stress \( \sigma_1 = 6 \) to 40 kPa) [180].

### 3.6.2 Elastic particle impact and maximum impact force

In terms of rapid collisional flow of particles within a shear zone, when two particles 1 and 2 approach and collide, their velocities are reduced gradually. Part of the initial kinetic energy is radiated into both particles as elastic waves. Now the contact force reaches a maximum value (maximum de-acceleration) and the particle velocities are reduced to zero. The elastic deformation work is calculated as [2, 4]:

\[ W_{el} = \frac{2}{3} \cdot E^* \int_0^{h_k} \sqrt{r_{1,2} \cdot h_k^3} \, dh_k = \frac{4}{15} \cdot E^* \cdot \sqrt{r_{1,2} \cdot h_k^5} \] (123)

With the average of particle mass according to Eq.(121) the correlation between the normal components of the relative velocity \( (v_1 - v_2) \) and the centre approach \( h_k \) is obtained:

\[ (v_1 - v_2)^2 = \frac{8 \cdot E^*}{15 \cdot m_{1,2} \cdot \sqrt{r_{1,2} \cdot h_k^5}} \approx \frac{E^*}{5 \cdot \pi \cdot \rho_s} \left( \frac{h_k}{r_{1,2}} \right)^{5/2} \] (124)

The maximum centre approach \( h_{K,\text{max}} \) at deformation rate \( \dot{h}_k = 0 \) is \( h_{K,\text{max}} \propto \Delta v^{4/5} \)

\[ h_{K,\text{max}} = \left[ \frac{225 \cdot m_{1,2}^2 \cdot (v_1 - v_2)^4}{64 \cdot E^* \cdot r_{1,2}^2} \right]^{1/5} \] (125)

and consequently, with Eq. (7) the maximum impact force \( F_{N,\text{max}} \propto \Delta v^{6/5} \) results in [4, 108]:

\[ F_{N,\text{max}} = \left[ \frac{125 \cdot m_{1,2}^3 \cdot E^* \cdot r_{1,2} \cdot (v_1 - v_2)^6}{144} \right]^{1/5} \] (126)

In the recovery stage the stored elastic energy is released and converted into kinetic energy and the particle moves with the rebound velocity \( v_{1,R} \) into the opposite direction. The contact duration for both, compression and decompression phase, is according to Hertz [108]:

\[ T_k = 3,7846 \cdot \left[ \frac{m_{1,2}^2}{E^* \cdot r_{1,2} \cdot (v_1 - v_2)} \right]^{1/5} \] (127)

For the limestone powder one obtains about \( T_k = 16 \) ns at a low shear rate of \( v_1 = 2 \) mm/min as perfect elastic contribution of contact displacement. It is worth to note here that plastic or additional adhesion contributions result in much higher contact durations.
3.6.3 Coefficient of restitution at particle impact

The so-called impact number or coefficient of restitution $e = \frac{\hat{F}_{1,R}}{\hat{F}_1}$ indicates the impact force ratio of the contact decompression (restitution) phase after impact and the contact compression phase during impact, with $e = 0$ for perfect plastic, $0 < e < 1$ for elastic-plastic and $e = 1$ for perfect elastic behaviours [312, 55], see examples in [65, 129, 141, 150, 313, 317, 319 - 324]. Thus $e^2 < 1$ characterizes the energy dissipation ($W_{\text{diss}}$ is the inelastic deformation work of particle contact, $E_{\text{kin},1} = m_1 \cdot v_1^2 / 2$ is the kinetic energy of particle 1 before impact):

$$e^2 = \frac{E_{\text{kin},1} - W_{\text{diss}}}{E_{\text{kin},1}}$$

(128)

Thus, the coefficient of restitution expresses the deceleration of the particle after impact and rebound $v_{1,R}$ or the reduction of rebound height $h_{1,R}$ (loss of potential energy) at free fall test [312, 320 - 322]:

$$e = \frac{v_{1,R}}{v_1} = \sqrt{\frac{h_{1,R}}{h_1}}$$

(129)

In terms of elastic-plastic contact deformation the initial kinetic energy is mainly dissipated into plastic dislocations, distortions and heat.

For a simple example, Walton [145] used linear normal force-displacement equations for plastic load $F_N = k_{N,\text{load}} \cdot h_K$ and for elastic unload $F_N = k_{N,U} \cdot (h_K - h_{K,E})$ along a secant line $U - E$ without any contact adhesion, see Fig. 5 panel b) and Table 1. The characteristic load and unload stiffnesses are equivalent to $k_{N,\text{load}} = k_{N,\text{el-pl}}$, Eq. (108), and $k_{N,U}$, Eq. (115). During the compression phase the kinetic energy of the particle 1 is converted into the potential energy of contact deformation during load $W_{\text{kin},1} = W_{\text{pot,load}}$. Thus, Eq. (128) can be rewritten as (index Wa):

$$e^2_{\text{Wa}} = \frac{W_{\text{pot,load}} - W_{\text{diss}}}{W_{\text{pot,load}}} = \frac{W_{\text{pot,load}} - (W_{\text{pot,load}} - W_{\text{pot,U}})}{W_{\text{pot,load}}} = \frac{\int k_{N,U} \cdot (h_K - h_{K,E}) \, dh_K}{k_{N,U} \cdot h_{K,E} \int 0}$$

(130)

$$e_{\text{Wa}} = \sqrt{\frac{k_{N,U} \cdot (h_{K,U} - h_{K,E})^2}{k_{N,\text{load}} \cdot h_{K,U}^2}}$$

(131)

The intersection point $U$ of load and unload lines results in the stiffness and displacement ratios:

$$\frac{k_{N,\text{load}}}{k_{N,U}} = \frac{h_{K,U} - h_{K,E}}{h_{K,U}} = \frac{h_{K,U} - h_{K,A}}{h_{K,U} + \kappa \cdot h_{K,A}}$$

(132)

With Eqs. (131) and (132), a constant coefficient of restitution $e_{\text{Wa}}$ is obtained independent of the relative velocity of approach [43, 145] which decreases with increasing unload stiffness.
\[ e_{wa} = \sqrt{\frac{h_{K,U} - h_{K,E}}{h_{K,U}}} = \sqrt{\frac{k_{N,load}}{k_{N,U}}} \]  

(133)

For example, neglecting the essential adhesion effect during decompression phase, the ultrafine limestone powder \( d_{50} = 1.2 \mu m \) yields \( e_{wa} \approx 0.57 \). This rough approach Eq. (133) describes the plastic contact damping behaviour without adhesion. But if the work needed to load \( W_{pot,load} \), unload \( W_{pot,U} \), elongate \( W_{pot,A} \) and detach the adhesion contact \( W_A \) (work of adhesion) are considered, this coefficient of restitution \( e_A \) results in a simple relation (index A, \( e_A \approx 0.56 \) for limestone powder):

\[ e_A = \sqrt{\frac{W_{pot,U}}{W_{pot,load} + W_{pot,A} + W_A}} = \sqrt{\frac{h_{K,U} - h_{K,A}}{h_{K,U}}} \approx \sqrt{\frac{h_{K,U} - h_{K,A}}{h_{K,U}}} \]  

(134)

For the rapid flow \( (v > 1 \text{ m/s}) \) of coarse particles without adhesion, a velocity variable coefficient of restitution, i.e. dominant viscous damping, seems to be more realistic and fits better experimental tests, see Walton [145]. Thus, the unloading slope \( k_{N,U} \) is expressed as a linear function of the maximum normal force \( F_{N,U,max} \) achieved before unloading [145]:

\[ k_{N,U} = k_{N,load} + S_N \cdot F_{N,U,max} \]  

(135)

With the eigenfrequency of an arbitrary particle 1 \( \omega_{0,1} \) and its mass \( m_1 \)

\[ \omega_{0,1} = \sqrt{\frac{2 \cdot k_{N,load}}{m_1}} \]  

(136)

the coefficient of restitution is calculated by [145]:

\[ e_{wa}(v_1) = \sqrt{\frac{\omega_{0,1}}{S_N \cdot v_1 + \omega_{0,1}}} \]  

(137)

Moreover, if one uses the centre approach \( h_{K,f} \) of Eq. (52) the critical impact velocity \( v_{1,f} \) for incipient plastic yield (index f) is calculated from Eq. (124) as [92, 318, 319]:

\[ v_{1,f} = \left( \frac{\pi \cdot p_f}{E \cdot \delta} \right)^2 \cdot \sqrt{\frac{p_f}{5 \cdot p_s}} \]  

(138)

Thus, for elastic-plastic contact deformation the coefficient of restitution was found by Thornton (index Th) [150, 151] as:

\[ e_{Th} = \sqrt{6 \cdot \sqrt{\frac{3}{5}} \cdot \left( 1 - \frac{v_{1,f}^2}{6 \cdot v_1^2} \right) \cdot \left( \frac{v_{1,f}}{v_1} \right)^{1/4}} \]  

(139)

This equation satisfies the condition \( e = 1.0 \) when \( v_1 = v_{1,f} \). At high impact velocities \( v_1 \gg v_{1,f} \) and Eq. (139) becomes [150, 151]:
This velocity dependent coefficient of restitution, Eq. (139) was confirmed recently by Antonyuk [340] for the impact behaviours of various granules [266, 267].

### 3.6.4 Particle impact and adhesion, contact displacement and impact force response

In terms of particle adhesion inside of the contact zone, a certain probability of sticking has to be taken into account. For this case a critical velocity (index H) as the stick/bounce criterion was derived by Thornton [151] who used the JKR model:

\[
v_{I, H, Th} = \frac{1.871 \cdot F_{H, JKR}}{m_p} \cdot \left( \frac{3 \cdot F_{H, JKR}^2}{d \cdot E^*} \right)^{1/3}
\]  

(141)

For the rapid collisional flow of coarse particles, at an impact velocity \( v_1 > v_{I, H} \) particle bounce occurs and the coefficient of restitution is obtained as [151]:

\[
e_{Th, JKR} = \sqrt{1 - \frac{v_{I, H}^2}{v_1^2}}
\]  

(142)

Even if the impact velocity \( v_1 \) is ten times higher than the critical sticking velocity \( v_{I, H, Th} \) the coefficient of restitution is 0.995 [151]. But this statement is in contradiction to the practical experience in ultrafine cohesive powder handling and their tendency to stick, i.e. \( e \to 0 \).

The critical velocity \( v_{I, H} \) to stick or to adhere the particles with a plastic contact deformation was derived by Hiller (Index HL) [313]:

\[
v_{I, H, HL} = \left( \frac{1 - e^2}{e^2} \right)^{1/2} \cdot \frac{1}{d} \cdot \frac{C_{H, e}}{\pi \cdot a_{F, 0}^2 \cdot \sqrt{6 \cdot \rho_s \cdot \rho_f}}
\]  

(143)

This can be rearranged if one uses the dimensionless plastic repulsion coefficient \( \kappa_p \) according to Eq. (73) to obtain the following simple expression:

\[
v_{I, H, HL} = \left( \frac{1 - e^2}{e^2} \right)^{1/2} \cdot \frac{a_{F, 0}}{d} \cdot \kappa_p \cdot \sqrt{6 \cdot \rho_f \rho_s}
\]  

(144)

Unfortunately, Eq. (144) is inconsistent for sticking particle after impact, i.e. \( e = 0 \), and does not include the increase of “soft” contact flattening response \( h_K \) by increasing particle impact velocity \( v_1 \).

Because of these problems to quantify the critical sticking velocity of an adhesion contact a new model is derived here. The dominant energy absorption \( W_{diss} \) during particle impact stressing, beginning at any unloading point \( U \), is considered approximately as a trapezium shaped area between elastic-plastic yield limit \( F_N(h_K) \), Eq. (99), and adhesion limit \( F_{N, Z}(h_K) \), Eq. (112), at infinite unload stiffness \( k_{N, U} \to \infty \) in the force-displacement diagram of Fig. 16. With the contribu-
tion of the work of adhesion \( W_A \) to separate this contact from equilibrium centre separation \( a_F=0 \) to infinity, the energy balance gives (\( A_{K,U} = \pi r_{1,2} a_{K,U} \) is the unload contact area):

\[
\frac{m_{1,2}}{2} \left( v_1^2 - v_{1,R}^2 \right) = \int_{h_{K,f}}^{h_{K,U}} F_N(h_K) \, dh_K + \int_{h_{K,f}}^{h_{K,U}} - F_{N,z}(h_K) \, dh_K + A_{K,U} \cdot \int_{a_F=0}^{\infty} \rho (h_{K,f}) \, (h_{K,0} - h_{K,f}) \, da
\]

With the averaged particle mass according to Eq. (121) one obtains after integration:

\[
v_{1}^2 - v_{1,R}^2 = \frac{3 \cdot p_f}{8 \cdot \rho_s \cdot r_{1,2}^2} \left[ \kappa_A \cdot \left( h_{K,U}^2 - h_{K,f}^2 \right) + \kappa_p \cdot a_{K,U} \cdot a_F=0 \right]
\]

Fig. 16: Back-calculated perfectly plastic contact deformation and sticking/bounce at central impact stressing using data from Fig. 12 for limestone. Two particles approach with velocities \( v_1 \) and \( v_2 \), impact and the contact is elastically-plastically deformed, upper panel. The inelastic deformation energy is dissipated into the contact. This is equivalent to the areas (grey tones) between elastic-plastic limit, Eq. (99), and adhesion limit, Eq. (112), for infinite unload stiffness which are obtained by integration, Eq. (145). If the kinetic energy of the particles would be large enough, these particles can detach with the rebound velocities \( v_{1,R} \) and \( v_{2,R} \). The critical impact velocity for incipient yield of the contact is shown, Eq. (138). Above this value, the two particles adhere or stick in practice, i.e. \( v_{1R} = 0 \). From this, the critical impact velocity \( v_{1,H} \) follows and is shown in the Figure below versus particle centre approach or displacement \( h_{K,U} \). The model of Hiller/Löffler (\( c \approx 0.5 \) assumed) predicts a constant velocity, Eq. (144). But the practical experience shows us that the faster the particles move and impact, the larger is the contact displacement, and as the consequence, the higher is the tendency to stick. This is demonstrated by the curve of Eq. (149) in the right panel versus displacement \( h_{K,U} \).

The difference in characteristic impact/rebound velocities results directly in the maximum centre approach, \( h_{K,U,max} \), expressed by the unloading point U:

\[
h_{K,U,max} = \frac{\kappa_p \cdot a_{F=0}}{2 \cdot \kappa_A} + \sqrt{\frac{(\kappa_p \cdot a_{F=0})^2}{2 \cdot \kappa_A} + \frac{8 \cdot \rho_s \cdot r_{1,2}^2}{3 \cdot \rho_f} \left( v_1^2 - v_{1,R}^2 \right) + h_{K,f}^2}
\]
The response of this displacement $h_{K,U,\text{max}}$ is a maximum contact consolidation force with nearly $F_{N,U,\text{max}} \propto v$:

$$F_{N,U,\text{max}} = \pi \cdot r_{1,2} \cdot p_f \cdot \left( \kappa_A - \kappa_p \right) \left[ \frac{\kappa_p \cdot a_{F=0}}{2 \cdot \kappa_A} \right]^2 + \frac{8 \cdot \rho_s \cdot r_{1,2}^2}{3 \cdot p_f} \left( v_{1,R} \right)^2 + h_{K,f}^2 - \frac{\kappa_p \cdot a_{F=0}}{2 \cdot \kappa_A} - F_{H0}$$

(148)

Additionally, a certain preconsolidation level, $F_{N,U}$, in a shear zone may affect the sticking/bounce probability. If the rebound velocity $v_{1,R} = 0$, the two particles will adhere, i.e. coefficient of restitution $e = 0$. Consequently, the critical sticking velocity $v_{1,H}$ is obtained without any additional losses, e.g. due to elastic wave propagation in solids by combination of compression (longitudinal), shear (transversal) and surface waves:

$$v_{1,H} = \sqrt{\frac{3 \cdot p_f}{8 \cdot \rho_s \cdot r_{1,2}^2} \left[ \kappa_A \cdot \left( h_{K,U}^2 - h_{K,f}^2 \right) + \kappa_p \cdot h_{K,U} \cdot a_{F=0} \right]}$$

(149)

For example using data from [180], this critical sticking velocity lies between 0.1 and 0.4 m/s for limestone, curve in the sticking velocity-displacement diagram of Fig. 16, which is equivalent to the macroscopic average pressures $\sigma_{M,st} = 3$ to 25 kPa. These calculation results of particle adhesion are in agreement with the practical experiences in powder handling and transportation, e.g. undesired powder blocking at conveyor transfer chutes. In terms of powder flow, the behaviour after multiple stressing of soft deforming contacts in the nanoscale can be approached as “healing contacts”, i.e., the contact deformation history $h_{K,U}$ is assumed to be “forgotten”.

To demonstrate this enormous adhesion potential, 1 µm silica particles were completely removed from a 100 mesh woven metal screen (147 µm wide) with 40 m/s air velocity [314] and 32 µm glass beads from glass surface with more than 117 m/s [316]. Air velocities of 10 to 20 m/s were necessary to blow off about 50% of quartz particles (d = 5 to 15 µm) which had adhered to filter media after impact velocities of about 0.28 to 0.84 m/s [64, 65].

These fundamentals of particle adhesion dynamics may also be important to chemically clean silicon wafers [87, 93 to 95, 259] or mechanical tool surfaces by jet pressures up to 2 MPa and CO$_2$-ice particle velocities up to 280 m/s [94].

### 3.7 Representative, load dependent adhesion force

Starting with these characteristic force-displacement functions one turns to a representative adhesion and normal force correlation to find out the physical basis of strength-stress relations in powder mechanics [18, 19, 82, 83, 98, 271, 185].

#### 3.7.1 Linear adhesion force – normal force function

*Instantaneous consolidation of representative contact*
Provided that the unloading curve is very stiff, one can directly replace the contact area in Eq. (68) for varying adhesion force by Eq. (94) and the following adhesion force - normal force relation is obtained:

\[
F_H = F_{H0} + p_{vaw} \cdot A_K = F_{H0} + \frac{p_{vaw}}{p_f} \frac{F_{H0} + F_N}{2 + \frac{1}{3} \frac{A_{pl}}{A_K} - \frac{p_{vaw}}{p_f}}
\]  \tag{150}

Therefore, with a so-called elastic-plastic contact consolidation coefficient \(\kappa\),

\[
\kappa = \frac{\kappa_p}{\kappa_A - \kappa_p}
\]  \tag{151}

a linear model for the adhesion force \(F_H\) as function of normal force \(F_N\) is obtained, see Tomas (1999/2000) [152, 153], Fig. 17:

\[
F_H = \frac{\kappa_A}{\kappa_A - \kappa_p} F_{H0} + \frac{\kappa_p}{\kappa_A - \kappa_p} F_N = (1 + \kappa) F_{H0} + \kappa F_N
\]  \tag{152}

The dimensionless strain characteristic \(\kappa\) is given by the slope of adhesion force \(F_H\) which is influenced by predominant plastic contact failure. It is a measure of irreversible particle contact stiffness or softness. A shallow slope designates low adhesion level \(F_H \approx F_{H0}\) because of stiff particle contacts, but a large slope means soft contacts, or consequently, cohesive powder flow behaviours [154, 178, 271].

Consequently, substituting the displacement \(h_K\) in Eqs. (114) and (116) results in a linear adhesion force model \(F_H(F_N)\) as well [339]. This function could be expressed either by the ratios of stiffness of the elastic-plastic yield limit \(k_{N,el-pl}\), the adhesion limit \(k_{N,A}\) and the secant line for unload \(k_{N,U}\), Eqs. (108), (113) and (115)

\[
F_H = \frac{1 + k_{N,A}/k_{N,el-pl}}{1 + k_{N,A}/k_{N,U}} F_{H0} + \frac{k_{N,U}/k_{N,el-pl} - 1}{k_{N,U}/k_{N,A} + 1} F_N
\]  \tag{153}

or, equivalent to this, by the elastic-plastic contact consolidation coefficients \(\kappa\) and \(\kappa_{Lu}\):

\[
F_H = \left(1 + \kappa \cdot \frac{h_{K,A}}{h_{K,U}}\right) F_{H0} + \kappa \cdot \frac{h_{K,A}}{h_{K,U}} F_N = (1 + \kappa_{Lu}) F_{H0} + \kappa_{Lu} F_N
\]  \tag{154}

This constitutive equation (154) describes additionally the history dependent pull-off force as a function of unload stiffness. Thus, the supplemented elastic-plastic contact consolidation coefficient \(\kappa_{Lu}\) is smaller than the slope \(\kappa\) in Eq. (152) (the index Lu relates to the linear force – displacement models of Luding [194] preferred to use in DEM simulations of cohesive powder flow dynamics [338, 339]):

\[
\kappa_{Lu} = \kappa \cdot \frac{h_{K,A}}{h_{K,U}}
\]  \tag{155}

But if the unload stiffness \(k_{N,U} \to \infty\) is infinite, i.e. \(h_{K,U} \approx h_{K,A}\), equivalence \(\kappa_{Lu} \approx \kappa\) of both elastic-plastic contact consolidation coefficients is obtained.
**Time consolidation of representative contact**

An elastic-plastic contact may be additionally deformed during the indentation time, e.g., by visco-plastic flow. The contact flattening is additionally dependent on time or displacement rate, section 3.2.5. Thus the contact reacts softer and, consequently, the adhesion level is higher than before and the adhesion force increases with interaction time [23, 57, 166]. Thus, the time-dependent consolidation behavior of particle contacts in a powder bulk was previously described by a parallel series, i.e. summation, of adhesion forces [98, 152, 269, 270]. This previous method refers more to incipient sintering or contact fusion of a thermally sensitive particle material, Rumpf [166], without interstitial adsorption layers.

Additionally, the increasing adhesion may be considered in terms of a sequence of rheological models as the sum of resistances due to plastic and visco-plastic repulsion $\kappa_p + \kappa_{p,t}$, 4th line in Table 3. These are characterised by the micro-yield strength $p_r$, apparent contact viscosity and time $\eta/K$. Hence the repulsion effect of “cold” viscous flow of comparatively strongly-bonded adsorption layers on the particle surface is taken into consideration [181, 272], e.g. limestone powder: specific surface area $A_{S, m}$ = 9.2 m²/g, surface moisture of $X_W$ = 0.5 % analyzed by Karl-Fischer titration. Hence with the total visco-plastic contact consolidation coefficient $\kappa_{vis}$ the linear correlation between total adhesion force and normal force $F_{H0}(F_N)$ from Eq. (152) can be written as, Table 3 and Fig. 17, Tomas (2004) [185]:

$$F_{H0,tot} = \frac{\kappa_{A,t}}{\kappa_{A,t} - \kappa_p - \kappa_{p,t}} \cdot F_{H0} + \frac{\kappa_p + \kappa_{p,t}}{\kappa_{A,t} - \kappa_p - \kappa_{p,t}} \cdot F_N = \left(1 + \kappa_{vis}\right) \cdot F_{H0} + \kappa_{vis} \cdot F_N$$  \hspace{1cm} (156)

This so-called total visco-plastic contact consolidation coefficient $\kappa_{vis}$ includes both the elastic-plastic $\kappa_p$ and the visco-plastic contributions $\kappa_{p,t}$ of contact flattening:

$$\kappa_{vis} = \frac{\kappa_p + \kappa_{p,t}}{\kappa_{A,t} - \kappa_p - \kappa_{p,t}}$$  \hspace{1cm} (157)

Eqs. (152) and (156) consider also the flattening response of soft particle contacts at normal force $F_N = 0$ caused by the adhesion force $\kappa \cdot F_{H0}$ (Krupp [142]) and $\kappa_{vis} \cdot F_{H0}$. Hence, the adhesion force $F_{H0}$ represents the sphere-sphere contact without any contact deformation at minimum particle-surface separation $a_F=0$.

### 3.7.2 Adhesion force of a stiff contact with surface asperity

This initial adhesion force $F_{H0}$ may also include a characteristic nanometre sized height or radius of a rigid spherical asperity $a_F=0 < h_s < r_1,2$ with its centre located at an average radius of the spherical particle, Krupp [57], Rumpf [78] and Schubert [80]:

$$F_{H0} = \frac{C_{H,als} \cdot h_s}{12 \cdot a_F} \cdot \frac{d / h_s}{\left[1 + \frac{d / h_s}{2 \cdot \left(1 + h_s / a_F\right)^2}\right]^2}$$  \hspace{1cm} (158)
This rigid adhesion force contribution $F_{H0}$ is valid only for perfect stiff contacts, see the fundamentals [111, 210, 211] and a number of supplements [57, 54, 75, 142, 217, 289 to 303]. According to Fig. 4b) in section 2.2, with the first derivative

$$\left.\frac{dF_{H0}}{dh}\right|_{dh=const} = 0$$

(159)

a minimum adhesion force $F_{H0,min}$ is obtained at the resulting roughness $h_{r,min}$:

$$h_{r,min} = \sqrt[3]{4 \cdot r_{1,2} \cdot a_{f=0}^2 - a_{f=0}}$$

(160)

$$F_{H0,min} \approx \frac{C_{H,lab} \cdot h_{r,min}}{12 \cdot a_{f=0}^2} \approx \left(\frac{4 \cdot r_{1,2}}{a_{f=0}}\right)^{\frac{1}{3}}$$

(161)

This reduction of adhesion force $F_{H0,min}$ can be used to improve the powder flowability in handling by coating the surface of these cohesive particles by nanoparticles [307]. The characteristic separation between these cohesive particles in a packing should be adjusted so that the 2nd complete moment of the roughness height distribution $Q_0(h_r)$ is equivalent to an averaged particle size $d_A$ of the nanosized flow additive. A square is assumed with $d_A^2$ as characteristic coverage area of the spherical nanoparticles. Therefore, the number of nanoparticles $n_A$ necessary to form a perfect monolayer at the surface of the cohesive particles according to their frequency distribution $q_0(d)$ is calculated as ($d_S$ surface diameter):

$$n_A = \frac{A_S}{A_{S,m}} = \frac{\pi \cdot \int d^2 \cdot q_0(d) \cdot d(d)}{h_{r,max} \cdot q_0(h_r) \cdot dh_r} \approx \frac{\pi \cdot d_S^2}{d_A^2}$$

(162)

With the 2nd and 3rd complete moments of particle size and nanoparticle size distributions $M_{2,0}$, $M_{3,0}$ one obtains ($A_{S,m}$ specific surface area):

$$\mu_A = \frac{n_A \cdot m_A}{m} = \frac{A_{S,m}}{A_{S,m,A}} = \frac{\pi \cdot M_{2,0}(d) \cdot \rho_{s,A} \cdot M_{3,0}(d_A)}{\rho_s \cdot M_{3,0}(d_A) \cdot M_{2,0}(d_A)} = \frac{\pi \cdot \rho_{s,A} \cdot \int d^2 \cdot q_0(d) \cdot d(d)}{d_A \cdot \int d^2 \cdot q_0(d_A) \cdot d(d_A)}$$

(163)

The mass fraction $\mu_A$ of flow additive of nanoparticles in a cohesive powder packing is estimated with Eq. (160) as:

$$\mu_A \approx \frac{\pi \cdot \rho_{s,A} \cdot h_{r,min}}{\rho_s \cdot d_S} \approx \frac{\pi \cdot \rho_{s,A} \left(\frac{a_{f=0}^2}{2 \cdot r_{1,2}}\right)^{\frac{1}{3}}}{2 \cdot \rho_s}$$

(164)

For example, a mass fraction of about $\mu_A = 1.5 \%$ is obtained to reduce the characteristic adhesion force $F_{H0}$ and improve the flow behaviour of the cohesive limestone powder.
Additionally, for rough spheres with diameter \( d = 4 \cdot r_{1,2} \) an averaged asperity height \( h_r = 2 \cdot h_{r1,2} \) can be used with \( h_{r1,2} = (1/h_r + 1/h_{r2})^{-1} \). When the asperity height is not too far from the averaged sphere radius \( h_{r1,2} < r_{1,2} \), then the adhesion force can be calculated as, Hoffmann (index Ho) [217]:

\[
F_{H0,H0} = \frac{C_{H,sls} \cdot h_{r1,2}}{6 \cdot a_{F=0}^2} \left[ \frac{1}{1 + h_{r1,2} / r_{1,2}} + \frac{r_{1,2} / h_{r1,2}}{(1 + 2 \cdot h_{r1,2} / a_{F=0})^2} \right] \approx \frac{C_{H,sls} \cdot h_{r1,2}}{6 \cdot a_{F=0}^2 \cdot (1 + h_{r1,2} / r_{1,2})} \quad (165)
\]

If the radius of the roughness exceeds the minimum separation of the sphere-plate system in order of magnitudes \( h_r >> a_{F=0} \), the contribution of the plate, second term in Eq. (165), can be neglected and the adhesion force may be described as the sphere-sphere contact [217].

Rabinovich et al. [304 - 306] have used the root mean square of roughness profile \( h_{r,RMS} \) from AFM measurements and the average peak to peak distance between these asperities \( \lambda_r \) to calculate the interaction between a smooth sphere and the surface with nanoscale roughness profile:

\[
F_{H0,Ra} = \frac{C_{H,sls} \cdot r_{1,2}}{6 \cdot a_{F=0}^2} \left[ \frac{1}{1 + 58.14 \cdot r_{1,2} \cdot h_{r,RMS} / \lambda_r^2} + \frac{1}{(1 + 1.817 \cdot h_{r,RMS} / a_{F=0})^2} \right] \quad (166)
\]

The first term in brackets represents the contact interaction of the particle with an asperity and the second term accounts for the non-contact interaction of the particle with an average surface plane. This approach describes stiff nanoscale roughness as caps of asperities with their centres located far below the surface. For example, roughness \( h_{r,RMS} \) of only 1 or 2 nm is significant enough to reduce the theoretical adhesion force \( F_{H0} \) by an order of magnitude or more [306]. Greenwood [125, 219, 220] described the elastic and plastic deformations of random surface asperities of contacts by the standard deviation of roughness and mean pressure.

The intersection of function (152) with abscissa \( F_H = 0 \) in the negative of consolidation force \( F_N \), Fig. 17, is surprisingly independent of the Hamaker constant \( C_{H,sls} \):

\[
F_{N,Z} = -\frac{\pi}{2} \cdot a_{F=0} \cdot h_r \cdot p_f \cdot \kappa_A \left[ 1 + \frac{d / h_r}{2 \cdot (1 + h_r / a_{F=0})^2} \right] \approx -\frac{\pi}{2} \cdot a_{F=0} \cdot h_r \cdot p_f \quad (167)
\]

This minimum normal (tensile or pull-off) force limit \( F_{N,Z} \) for nearly brittle contact failure combines the influences micro-yield strength or stiffness of the particle contact and the particle separation distribution, which is characterised here by the mean particle roughness height \( h_{r1,2} \), and the molecular centre separation \( a_{F=0} \). Obviously, this value \( F_{N,Z} \) characterises also the effective \textit{contact softness} with respect to asperity displacement and their curve radius \( h_{r1,2} \) before flattening, see Eq. (58) in section 3.2.5.

This elastic-plastic model Eq. (152) can be interpreted as a general linear constitutive contact model concerning loading pre-history dependent particle adhesion, i.e. linear in forces and stresses, but non-linear regarding material characteristics.
3.7.3 Non-linear adhesion force – normal force function

In section 3.3 the non-linear Johnson-Maugis-Pollock model Eq. (75) and its extension by Castellanos, Eq. (77), are explained. To compare these non-linear models with those described in the above section, one replaces the surface energy $\sigma_{sls}$ in Eq. (75) by the plastic repulsion coefficient $\kappa_p$ with Eq. (73). An equivalent adhesion force - normal force function is obtained and used here with a fit factor of 1/3, see the dashed-double dotted brown curve in Fig. 17, to compare it with the representative adhesion forces back-calculated from the powder shear tests:

$$F_{h} = \frac{\kappa_p \cdot a_{f=0} \cdot E^*}{6} \cdot \sqrt{\frac{F_N + F_{h0}}{\pi \cdot p_f}}$$ (168)

Moreover, if one eliminates the centre approach $h_K$ of the loading and unloading functions, Eqs. (99) and (104), an implied non-linear function between the contact pull-off force $F_{h} = - F_{N,Z}$ at the detachment point A is obtained for the normal force at the unloading point $F_N = F_{N,U}$:

$$F_{h(t)} = F_{h0} + \kappa \cdot (F_N + F_{h0}) - \pi \cdot a_{f=0} \cdot \kappa_p \cdot p_f \cdot \left[ \frac{3 \cdot (F_N + F_{h0})}{2 \cdot a_{f=0} \cdot E^*} \cdot \left( 1 + \frac{F_{h(t)=0} - F_{h0}}{F_N + F_{h0}} \right) \right]^{2/3}$$ (169)

Fig. 17: Representative particle adhesion forces for cohesive limestone powder (median $d_{50,3}$ = 1.2 $\mu$m) according to the linear Eq. (152) with the elastic-plastic contact consolidation coefficient $\kappa = 0.224$ (red dashed-dotted line), linear Eq. (154) with $\kappa_{LU} = 0.14$ (green dotted line), implied non-linear Eq. (169) (blue dotted curve), the dashed-double dotted brown curve of non-linear model Eq. (168), all for contact time $t \approx 0$, i.e. instantaneous consolidation, and the linear model for time consolidation $t = 24$ h Eq. (156) with the total viscoplastic contact consolidation coefficient $\kappa_{vis} = 1.014$ (black line). The powder surface moisture $X_W = 0.5 \%$ (specific surface area $A_{S,m} = 9.2$ m$^2$/g, accurately analyzed by Karl-Fischer titration) equals idealized bi-molecular adsorption layers being in equilibrium with ambient air of 20°C temperature and 50% humidity.
This unloading point \( U \) is stored in the memory of the contact as preconsolidation history. This general non-linear adhesion model, dashed blue curve in Fig. 17, implies the dimensionless, elastic-plastic contact consolidation coefficient \( \kappa \) and, additionally, the influence of adhesion, stiffness, average particle radius \( r_{1,2} \), average modulus of elasticity \( E^* \) in the last term of the equation. The sphere-sphere interaction without any contact deformation \( F_{H0} \) and the non-linear adhesion force contribution are influenced by the radius of non-deformed surface curvature \( r_{1,2} \) or particle size \( d \), see Tomas (2004) [184, 185].

It is good to see that in Fig. 17 all curves (for contact time \( t \approx 0 \)) are found to be in the same range. Thus, the linearised adhesion force equation (152) is used first to demonstrate comfortably the correlation between the adhesion forces of microscopic particles and the macroscopic stresses in powders [152, 154, 178]. The micromechanical elastic-plastic contact consolidation correlates directly with the macro-mechanical powder flowability expressed by the semi-empirical flow function \( f_{fc} \) according to Jenike [7], see Tomas [155, 271].

![Fig. 18: The influence of particle size \( d \) on adhesion force \( F_H \) according to the linear model, Eq. (152), and non-linear model Eq. (169) with normal force \( F_N \) as curve parameter, calculated with the coefficients of Fig. 17. For ultrafine particles with soft contacts (about \( d < 1 \) \( \mu m \)) the adhesion force dominates mainly by the size-independent contribution of normal force. For coarse particles with stiff contacts (\( d > 50 \) \( \mu m \)) the adhesion force becomes more and more independently of normal force and the contribution of \( F_{H0} \) dominates, see Fig. 4 panel a).](image)

It should be pointed out here that the adhesion force level in Fig. 18 is approximately \( 10 - 10^6 \) times the particle weight for fine particles \( d < 100 \) \( \mu m \), see the dashed-dotted straight-line marked with FG. This means, in other words, that one has to apply these large values as acceleration ratios \( a/g \) with respect to gravity to separate these pre-consolidated contacts or to remove mechanically such adhered particles from surfaces.

For moist particle packings, the liquid bridge bonding forces caused by capillary pressure of interstitial pores and surface tension contribution of the free liquid surface additionally determine the strength [96, 98, 308]. Capillary pressure and increasing van der Waals adhesion forces by
contact flattening due to normal load attract also particle contacts of compressed water-saturated filter cakes or wet-mass powders [178, 272, 332 - 336]. Macroscopic pressures from 50 kPa to 10 MPa are used to form, compress and consolidate filter cakes [333]. Despite these large pressures, the microscopic normal stresses are limited by the yield limit \( p_f \) of the contact, Fig. 8. Generally, the bonds within the solid matrix are more intensive and stronger than bonds between the ions of particle surfaces, electrostatic double layers and van der Waals forces of polar fluid molecules. Under constraints of compressed particle packing theses repulsive double layers [58, 326] are displaced within the contact. Direct contacts are formed with contact flattening and load dependent adhesion as consequence. This leads to macroscopic consolidation of a few hundred kilopascals or more in a compressed and drained filter cake [332 - 334] equivalent to that what happens in dry or humid environment. Coulomb friction is activitated between these contact plates when the elastic or yield limit is reached at comparatively low tangential contact displacements \( \delta > \delta_0 \approx 1 \text{ nm} \).

3.7.4 Elastic-plastic, frictional tangential force of load dependent adhesion contact

The tangential stiffness according to Mindlin [117], Eq. (19), can be written for the elastic-plastic contact flattening caused by a sufficiently large normal force \( F_N \) and, consequently, partially sticking by load dependent adhesion force \( F_H(F_N) \), Eq. (152), as function of tangential displacement \( \delta \) (index H for adhesive contact):

\[
k_{T,H} = \frac{dF_T}{d\delta} = 4 \cdot G \cdot r_K \cdot \left( 1 - \frac{\delta}{\delta_{c,H}} \right)^{1/2}
\]

The initial stiffness \( k_{T,H0} \) is found at a tangential displacement \( \delta = 0 \):

\[
k_{T,H0} = \left. \frac{dF_T}{d\delta} \right|_{\delta=0} = 4 \cdot G \cdot r_K = 4 \cdot G \cdot \sqrt{r_{t,2} \cdot h_K}
\]

Thus, the tangential force-displacement relation can be expressed by a linear elastic contribution for the no-slip region within the contact area according to Eq. (20), Hook’s law, Fig. 6 panel a):

\[
F_t = 4 \cdot G \cdot r_K \cdot \delta
\]

The contact radius \( r_K \) depends on the actual normal force \( F_N \) of the elastic-plastic yield limit Eq. (96) in normal direction and on the effective adhesion force \( F_H(F_N) \), Eq. (152)

\[
F_t = 4 \cdot G \cdot \frac{(1 + \kappa) \cdot (F_{H0} + F_N)}{\pi \cdot \kappa_\lambda \cdot p_t} \cdot \delta,
\]

so that the initial tangential stiffness \( k_{T,H0}(F_N) \), Eq. (171), increases with increasing preconsolidation \( F_H(F_N) \), see Fig. 19. This stiffness amounts to \( k_{T,H0} = 800 - 2000 \text{ N/m} \) for representative limestone particles.

The Coulomb friction limit of the tangential force is described by the coefficient of internal friction \( \mu_i \) and depends on the elastic-plastic contact consolidation, i.e., elastic-plastic flattening by
the given normal force $F_N$ and the variable adhesion force $F_{H}(F_N)$ as well, see Eqs. (152) or (169) in the previous sections 3.7.1 and 3.7.3:

$$F_{T,H} = \mu_i \cdot [F_N + F_H(F_N)] = \mu_i \cdot (1 + \kappa) \cdot (F_{H0} + F_N)$$  \hspace{1cm} (174)

Using Eqs. (96), (172) and (174) the transition between the linear elastic range and incipient contact failure by Coulomb friction is calculated by the friction limit of tangential displacement $\delta_{0,H}$, see the red points for yield in Fig. 19:

$$\delta_{0,H} = \frac{\mu_i \cdot [F_N + F_H(F_N)]}{4 \cdot G \cdot r_K(F_N)} = \frac{\mu_i}{4 \cdot G} \cdot \sqrt{\pi \cdot \kappa_A \cdot p_f \cdot (1 + \kappa) \cdot (F_{H0} + F_N)}$$  \hspace{1cm} (175)

Moreover, with the Coulomb friction limit, Eq. (18), the contact loses his elastic tangential stiffness at $k_{T,H} = 0$ and completely mobilized contact sliding is obtained for the new friction limit of displacement $\delta = \delta_{C,H}$ under load dependent adhesion force $F_{H}(F_N)$, Eq. (152):

$$\delta_{C,H} = \frac{3 \cdot \mu_i \cdot (1 + \kappa) \cdot (F_{H0} + F_N)}{8 \cdot G \cdot r_K} = \frac{3 \cdot \mu_i}{8 \cdot G} \cdot \sqrt{\pi \cdot \kappa_A \cdot p_f \cdot (1 + \kappa) \cdot (F_{H0} + F_N)},$$  \hspace{1cm} (176)

Both friction limits, Eqs. (175) and (176) are similar $\delta_{C,H} = 1.5 \cdot \delta_{0,H}$ and increase with increasing load $F_N$. Thus Mindlin’s nonlinear model $F_T = f(\delta)$ is supplemented for first loading, Eq. (14):
\[ F_T = F_{T,C,H} \left(1 - \left(1 - \frac{\delta}{\delta_{C,H}}\right)^{3/2}\right), \]  

(177)

Therefore, the functions for unloading and shear in reverse direction (- sign) according to Mindlin and Deresiewicz [118], Eq. (23), are

\[ F_{T,U} = F_{T,U}^{\text{r}} - 2 \cdot F_{T,C,H} \cdot \left[1 - \left(1 - \frac{\delta_U - \delta}{2 \cdot \delta_{C,H}}\right)^{3/2}\right], \]  

(178)

and for reloading in the previous (+) shear direction, Eq. (24) for \( F_{T,\text{reload}} = -F_{T,U} \) and \( \delta_{\text{reload}} = -\delta_U \):

\[ F_{T,\text{reload}} = -F_{T,\text{reload}}^{\text{r}} + 2 \cdot F_{T,C,H} \cdot \left[1 - \left(1 - \frac{\delta + \delta_{\text{reload}}}{2 \cdot \delta_{C,H}}\right)^{3/2}\right], \]  

(179)

The essential force – displacement models \( F_T(\delta) \) models are collected in Table 4:

<table>
<thead>
<tr>
<th>Process</th>
<th>Model</th>
<th>Equation</th>
<th>Eq.</th>
</tr>
</thead>
<tbody>
<tr>
<td>stiffness</td>
<td>load dependent adhesion</td>
<td>[ k_{T,H} = \frac{dF_T}{d\delta} = 4 \cdot G \cdot \kappa \cdot \left(1 - \frac{\delta}{\delta_{C,H}}\right)^{1/2} ]</td>
<td>(170)</td>
</tr>
<tr>
<td>Initial stiffness</td>
<td>load dependent</td>
<td>[ k_{T,H_0} = \frac{dF_T}{d\delta}\bigg</td>
<td><em>{\delta=0} = 4 \cdot G \cdot \kappa \cdot \sqrt{r</em>{1,2} \cdot h_k} ]</td>
</tr>
<tr>
<td>Friction limits</td>
<td>tangential force displacement</td>
<td>[ F_{T,C,H} = \mu_i \cdot (1 + \kappa) \cdot (F_{H_0} + F_N) ]</td>
<td>(174)</td>
</tr>
<tr>
<td>Loading</td>
<td>0 \leq \delta \leq \delta_{C,H}</td>
<td>[ F_T = F_{T,C,H} \cdot \left[1 - \left(1 - \frac{\delta}{\delta_{C,H}}\right)^{3/2}\right] ]</td>
<td>(177)</td>
</tr>
<tr>
<td>Unloading</td>
<td>-\delta_{C,H} \leq \delta \leq \delta_{C,H}</td>
<td>[ F_T = F_{T,U} - 2 \cdot F_{T,C,H} \cdot \left[1 - \left(1 - \frac{\delta_U - \delta}{2 \cdot \delta_{C,H}}\right)^{3/2}\right] ]</td>
<td>(178)</td>
</tr>
<tr>
<td>Reloading</td>
<td>-\delta_{C,H} \leq \delta \leq \delta_{C,H}</td>
<td>[ F_T = -F_{T,\text{reload}} + 2 \cdot F_{T,C,H} \cdot \left[1 - \left(1 - \frac{\delta + \delta_{\text{reload}}}{2 \cdot \delta_{C,H}}\right)^{3/2}\right] ]</td>
<td>(179)</td>
</tr>
</tbody>
</table>

Beginning with this displacement limit \( \delta_{0,H} \), energy is mainly dissipated by irreversible friction work. The particle mass related (specific) friction work \( W_{m,T} \) results in (k coordination number):

\[ W_{m,T} = k \int_{\delta_{0,H}}^{\delta} F_T(\delta^*) \, d\delta^* \]  

(180)
\[ W_{m,T} = \frac{k \cdot F_{T,C,H} \cdot (\delta - \delta_{0,H})}{m_{1,2}} = \frac{k \cdot \mu_i \cdot (1 + \kappa) \cdot (F_{H0} + F_N) \cdot (\delta - \delta_{0,H})}{m_{1,2}} \] (181)

If one assumes a characteristic displacement of about one contact radius \((\delta - \delta_{0,H}) \approx r_K\) this mass related friction work amounts to 1.0 - 13 mJ/g for the limestone particles at different preconsolidation levels of average pressures \(\sigma_{M,st} = 3\) to 25 kPa of the bulk powder.

The dissipated mechanical work per one closed cycle between unload \(\delta_U\) and reload \(\delta_{\text{reload}}\) displacements according to Eq. (27) is obtained as:

\[ W_{T,H} = 8 \cdot F_{T,C,H} \cdot \delta_{C,H} \left\{ \left( 1 - \frac{F_{\text{reload}}^* + F_{\text{U}}^*}{4 \cdot F_{T,C,H}} \right) \cdot \delta_{U} + \delta_{\text{reload}} \right\} \left\{ 2 \cdot \delta_{C,H} \right\} \frac{1}{5} \left[ 1 - \left( 1 - \frac{\delta_{U} + \delta_{\text{reload}}}{2 \cdot \delta_{C,H}} \right)^{5/2} \right] \] (182)

For the characteristic displacements between these reload/unload limits due to Coulomb friction \(F_{\text{reload}}^* = -F_{T,C,H}\) and \(F_{\text{U}}^* = F_{T,C,H}\) at \(\delta_{\text{reload}} = -\delta_{C,H}\) and \(\delta_U = \delta_{C,H}\) the maximum mass related hysteresis work or specific energy absorption for one cycle results in (k coordination number, \(m_{1,2}\) average particle mass according to Eq. (120)):

\[ W_{m,T,H,\text{max}} = \frac{4 \cdot k}{5 \cdot m_{1,2}} \cdot F_{T,C,H} \cdot \delta_{C,H} \] (183)

This work amounts only to 0.6 – 8.0 \(\mu\)J/g compared to the large 1.0 - 13 mJ/g for the specific friction work at \(\delta \approx r_K = 6 - 14\) nm, Eq. (181), of limestone particles at different macroscopic preconsolidation levels of average pressures \(\sigma_{M,st} = 3\) to 25 kPa of the bulk powder.

### 3.7.5 Elastic-plastic, frictional rolling resistance of load dependent adhesion contact

Using Johnson’s creep model Eq. (28), supplemented, rearranged and differentiated, and the stiffness \(k_{R,H}\) of rolling resistance \(F_R\) by elastic-plastic contact flattening and consequently, partially sticking, follows as function of relative rolling angle \(\gamma\) of both spheres (index H for adhesion):

\[ k_{R,H} = \frac{dF_R}{d\gamma} = \frac{16 \cdot G}{\pi \cdot (4 - 3 \cdot v)} \left( 1 + \kappa \right) \left( F_{H0} + F_N \right) \frac{1 - \frac{\gamma}{\gamma_{C,H}}}{\kappa_A \cdot p_f} \] (184)

\[ \gamma = \tan \left( \frac{\delta_1}{r_1} \right) - \tan \left( \frac{\delta_2}{r_2} \right) \approx \frac{\delta_1}{r_1} - \frac{\delta_2}{r_2} \] (185)

The initial stiffness \(k_{R,H0}\) is found at zero of rolling angle \(\gamma = 0\):

\[ k_{R,H0} = \left. \frac{dF_R}{d\gamma} \right|_{\gamma=0} = \frac{16 \cdot G}{\pi \cdot (4 - 3 \cdot v)} \left( 1 + \kappa \right) \left( F_{H0} + F_N \right) \frac{1}{\kappa_A \cdot p_f} \] (186)

The contact looses his elastic rolling stiffness at \(k_{R,H} = 0\) and the rolling friction limit with respect to a critical rolling angle \(\gamma = \gamma_{C,H}\) is obtained:

\[ \gamma_{C,H} = \frac{3 \cdot \alpha_R \cdot (4 - 3 \cdot v)}{16 \cdot G \cdot r_{1,2}} \cdot \sqrt{\pi \cdot \kappa_A \cdot p_f \cdot (1 + \kappa) \cdot (F_N + F_{H0})} \] (187)
With Eqs. (31) and (15) for elastic-plastic contact displacement in normal direction and load dependent adhesion force, by a tilting moment relation at the perimeter of flattened contact, Fig. 20 panel right, the critical rolling resistance $F_{R,C,H}$ results in:

$$F_{R,C,H} = \frac{r_K}{r} \cdot (1 + \kappa) \cdot (F_{H0} + F_N)$$  \hspace{1cm} (188)

Substitution of the contact radius $r_K$ of the elastic-plastic yield limit Eq. (96) results nearly in a proportionality as $F_{R,C,H}(F_N) \propto F_N^{3/2}$:

$$F_{R,C,H} = \mu_R \cdot (1 + \kappa) \cdot (F_{H0} + F_N) = \alpha_R \cdot \left( \frac{(1 + \kappa)^3 \cdot (F_{H0} + F_N)^3}{\pi \cdot r_{1/2}^3 \cdot p_r \cdot \kappa} \right)$$  \hspace{1cm} (189)

Fig. 20: Representative rolling resistance force - rolling angle diagram for limestone particles (median $d_{50,3} = 1.2 \mu m$, shear modulus $G^* = 34 \text{kN/mm}^2$, rolling friction coefficient $\mu_R \approx 0.01 - 0.024$). This frictional rolling resistance of smooth soft spheres can be considered by a tilting moment relation of the force pair around the red pivot at the perimeter of contact circle. As the response of contact flattening a lever arm of contact radius $r_K$ with respect to $F_N$ is generated which is equilibrated by rolling resistance $F_R$ acting perpendicular to direction of $F_N$ with the lever arm $r-h\kappa/2$, see the blue lines of panel right. The linear elastic range is very small and limited by the critical rolling angle $\gamma_{C,H} = 8 \times 10^{-5} - 1.9 \times 10^{-4}$. At this yield point the elastic behaviour is transmitted into the frictional behaviour of contact rolling shown by constant rolling resistance force, Eq. (189). The elastic-plastic contact flattening in normal direction and resulting load dependent adhesion force $F_{H}(F_N)$ is the parameter of this rolling friction limit as well.

It is advisable to distinguish between the load dependent coefficient of rolling friction

$$\mu_R(F_N) = \alpha_R \cdot \frac{r_K(F_N)}{r},$$  \hspace{1cm} (190)

and an adjustable load independent coefficient of rolling resistance $\alpha_R$, see Iwashita [232]. Thus, the micro-slip within the contact plane can be considered, i.e., with $\alpha_R = 1$ for free rolling with-
out micro-slip and $\alpha_R = \alpha_{R,\text{max}}$ for the fully developed tangential sliding $\mu_R = \mu_l$ at large contact deformations and $F_{R,C,H} = F_{T,C,H}$:

$$1 \leq \alpha_R \leq \alpha_{R,\text{max}} = \mu_l \cdot r / r_k$$  \hspace{1cm} (191)

By integration of the stiffness $k_{R,H}$, Eq. (184), the force – rolling angle function for the first loading is obtained:

$$F_R = F_{R,C,H} \cdot \left[ 1 - \left( 1 - \frac{\gamma}{\gamma_{C,H}} \right)^{\frac{3}{2}} \right]$$ \hspace{1cm} (192)

Similar to the constitutive tangential force - displacement model, Eqs. (178) and (179), the function for unloading and rolling in reverse direction (- sign) is obtained as

$$F_R = F_{R,U} - 2 \cdot F_{R,C,H} \cdot \left[ 1 - \left( 1 - \frac{\gamma_{U} - \gamma}{2 \cdot \gamma_{C,H}} \right)^{\frac{3}{2}} \right]$$ \hspace{1cm} (193)

and for reloading in the previous (+) rolling direction, i.e. $F_{R,\text{reload}} = -F_{R,U}$ and $\gamma_{\text{reload}} = -\gamma_{U}$:

$$F_R = -F_{R,\text{reload}} + 2 \cdot F_{R,C,H} \cdot \left[ 1 - \left( 1 - \frac{\gamma_{\text{reload}} + \gamma}{2 \cdot \gamma_{C,H}} \right)^{\frac{3}{2}} \right]$$ \hspace{1cm} (194)

These force – displacement models $F_R(\gamma)$ models are shown again in Table 5:

**Table 5:** Compilation of rolling resistance force – rolling angle $F_R(\gamma)$ of load dependent van der Waals adhesion for load, elastic-plastic contact deformation, unload and reload, see Fig. 20.

<table>
<thead>
<tr>
<th>Process</th>
<th>Model</th>
<th>Equation</th>
<th>Eq.</th>
</tr>
</thead>
<tbody>
<tr>
<td>stiffness</td>
<td>load dependent adhesion</td>
<td>$k_{R,H} = \frac{df_R}{d\gamma} = \frac{16 \cdot G}{\pi \cdot (4 - 3 \cdot v)} \cdot \frac{(1 + \kappa) \cdot (F_{H0} + F_N)}{\kappa_A \cdot p_f} \cdot \left( 1 - \frac{\gamma}{\gamma_{C,H}} \right)^2$</td>
<td>(184)</td>
</tr>
<tr>
<td>Initial stiffness</td>
<td>load dependent</td>
<td>$k_{R,H,0} = \frac{df_R}{d\gamma} \bigg</td>
<td><em>{\gamma=0} = \frac{16 \cdot G}{\pi \cdot (4 - 3 \cdot v)} \cdot \frac{(1 + \kappa) \cdot (F</em>{H0} + F_N)}{\kappa_A \cdot p_f}$</td>
</tr>
<tr>
<td>Friction limits $k_{R,H} = 0$</td>
<td>Rolling resistance force</td>
<td>$F_{R,C,H} = \alpha_R \cdot \frac{(1 + \kappa)^{\frac{3}{2}} \cdot (F_{H0} + F_N)}{\pi \cdot r_{t,2} \cdot p_f \cdot \kappa_A}$</td>
<td>(189)</td>
</tr>
<tr>
<td></td>
<td>Rolling angle</td>
<td>$\gamma_{C,H} = \frac{3 \cdot \alpha_R \cdot (4 - 3 \cdot v)}{16 \cdot G \cdot r_{t,2}} \cdot \sqrt{\pi \cdot \kappa_A \cdot p_f \cdot (1 + \kappa) \cdot (F_N + F_{H0})}$</td>
<td>(187)</td>
</tr>
<tr>
<td>Loading</td>
<td>rolling force – angle relation</td>
<td>$F_R = F_{R,C,H} \cdot \left[ 1 - \left( 1 - \frac{\gamma}{\gamma_{C,H}} \right)^{\frac{3}{2}} \right]$</td>
<td>(192)</td>
</tr>
<tr>
<td>$0 \leq \gamma \leq \gamma_{C,H}$</td>
<td>Unloading</td>
<td>$F_R = F_{R,U} - 2 \cdot F_{R,C,H} \cdot \left[ 1 - \left( 1 - \frac{\gamma_{U} - \gamma}{2 \cdot \gamma_{C,H}} \right)^{\frac{3}{2}} \right]$</td>
<td>(193)</td>
</tr>
<tr>
<td>Unloading</td>
<td>frictional behaviour</td>
<td>$F_R = F_{R,U} - 2 \cdot F_{R,C,H} \cdot \left[ 1 - \left( 1 - \frac{\gamma_{U} - \gamma}{2 \cdot \gamma_{C,H}} \right)^{\frac{3}{2}} \right]$</td>
<td>(193)</td>
</tr>
<tr>
<td>$-\gamma_{C,H} \leq \gamma \leq \gamma_{C,H}$</td>
<td>Reloading</td>
<td>$F_R = -F_{R,\text{reload}} + 2 \cdot F_{R,C,H} \cdot \left[ 1 - \left( 1 - \frac{\gamma_{\text{reload}} + \gamma}{2 \cdot \gamma_{C,H}} \right)^{\frac{3}{2}} \right]$</td>
<td>(194)</td>
</tr>
</tbody>
</table>
Beyond the rolling friction limit \( \gamma_{C,H} \), energy is dissipated by an irreversible work of the rolling resistance \( F_R \) or its rolling moment \( M_R = F_R r \). Thus, the particle mass related rolling resistance work \( W_{m,R} \) results in \((k\text{ coordination number})\):

\[
W_{m,R} = \frac{k}{m_{1,2}} \int_{\gamma_{C,H}}^{\gamma} M_R (\gamma') \, d\gamma' = \frac{k \cdot M_R \cdot (1 + \kappa) \cdot (F_{H0} + F_N) \cdot r_{1,2} \cdot (\gamma - \gamma_{C,H})}{m_{1,2}}
\]

(195)

### 3.7.6 Elastic-plastic, frictional twisting of load dependent adhesion contact

The torsional stiffness according to Deresiewicz, Eq. (33), can be written for elastic-plastic contact flattening and consequently, partially sticking by load dependent adhesion force \( F_{H}(F_N) \), Eq. (152), as function of torsional moment \( M_{to} \):

\[
k_{to,H} = \frac{dM_{to}}{d\phi} = \frac{8 \cdot G \cdot r_k^2}{3} \left[ 2 \cdot \left( 1 - \frac{M_{to}}{M_{to,C,H}} \right)^{-1/2} - 1 \right]^{-1}
\]

(196)

The initial stiffness \( k_{to,H0} \) is found at vanishing torsional moment \( M_{to} = 0 \):

\[
k_{to,H0} = 2 \cdot (1 - \nu) \cdot r_{1,2} \cdot (1 + \kappa) \cdot (F_{H0} + F_N)
\]

(197)

This Eq. (197) corresponds to the initial torsional stiffness of Mindlin, Eq. (35), supplemented by the load dependent adhesion force \( F_{H}(F_N) \). For for the limestone powder \( (d_{50} = 1.2 \, \mu m) \) it gives very small values \( k_{to,H} = (35 – 500) \times 10^{-15} \, \text{Nm} \). Thus for simplicity, the elastic moment - rotation angle function can be approached as linear relation, Fig. 21.

The contact loses his elastic torsional stiffness at \( k_{to,H} = 0 \) and completely mobilized “frictional” contact rotation is obtained for the friction limit of moment \( M_{to} = M_{to,C,H} \) under load dependent adhesion force \( F_{H}(F_N) \):

\[
M_{to,C,H} = \frac{2 \cdot \mu_1 \cdot (1 + \kappa) \cdot (F_{H0} + F_N) \cdot r_k}{3}
\]

(198)

If one integrates the elastic-frictional torsional stiffness \( k_{to,H} \), Eq. (196), the torsional moment \( M_{to} \) as function of relative rotation angle \( \phi = \phi_1 - \phi_2 \) is obtained with Eq. (152) for loading (unfortunately, Deresiewicz’ [40] Eqs. (17), (23) and (25) could not suitable applied for this case):

\[
M_{to} = 4 \cdot M_{to,C,H} \cdot \left[ \sqrt{\frac{3 \cdot \phi}{\phi_{C,H}} + 1} - \frac{3 \cdot \phi}{4 \cdot \phi_{C,H}} - 1 \right]
\]

(199)

With the moment of completely mobilized frictional contact rotation \( M_{to,C,H} \), Eq. (198), results in the critical rotation angle \( \phi_{C,H} \) under load dependent adhesion force \( F_{H}(F_N) \):

\[
\phi_{C,H} = \frac{3 \cdot \mu_1 \cdot (1 + \kappa) \cdot (F_{H0} + F_N)}{4 \cdot G \cdot r_k^2}
\]

(200)

Substituting contact radius \( r_k \) by Eq. (96) yields the simple constitutive relation for the frictional limit of rotation of an elastic-plastic contact with adhesion.
\[ \phi_{C,H} = \frac{3 \cdot \pi \cdot \mu_1 \cdot \kappa_\chi \cdot \rho_f}{4 \cdot G} \],

(201)

which depends only on the particle properties, but is independent of the load \( F_N \) (provided that \( \kappa_\chi = \text{const.} \)). This critical value amounts to \( \phi_{C,H} \approx 0.0076 \) for limestone powder, Fig. 21.

This new model for loading of an elastic-plastic frictional contact Eq. (199) may be supplemented by the models for unloading, i.e. twisting in the reverse direction,

\[ M_{to,unload} = M_{to,U} - 8 \cdot M_{to,C,H} \left[ \frac{3 \cdot (\phi_U - \phi)}{2 \cdot \phi_{C,H}} + 1 - \frac{3 \cdot (\phi_U - \phi)}{8 \cdot \phi_{C,H}} \right] \]

(202)

and for reloading, i.e. twisting in the previous direction:

\[ M_{to,reload} = -M_{to,U} + 8 \cdot M_{to,C,H} \left[ \frac{3 \cdot (\phi_U + \phi)}{2 \cdot \phi_{C,H}} + 1 - \frac{3 \cdot (\phi_U + \phi)}{8 \cdot \phi_{C,H}} \right] \]

(203)

Beyond the friction limit \( \phi_{C,H} \), energy is dissipated by irreversible torsional friction work. The particle mass related torsional friction work \( W_{m,to} \) results in (k coordination number):

\[ W_{m,to} = \frac{k}{m_{1,2}} \frac{\phi}{\phi_{C,H}} \int_{M_{10}(\phi^*)} d\phi^* = 2 \cdot k \cdot \mu_1 \cdot (1 + \kappa) \cdot (F_{H10} + F_N) \cdot r_k \cdot (\phi - \phi_{C,H}) \]

(204)

To make the survey and comparisons more convenient, the necessary equations for the elastic-plastic, frictional behaviours with load dependent adhesion are shown again in Table 6.
Table 6: Compilation of torsional moment – rotation angle models $M_{to}(\phi)$ of load dependent van der Waals adhesion for load, elastic-plastic contact deformation, unload and reload, see Fig. 21.

<table>
<thead>
<tr>
<th>Process</th>
<th>Model</th>
<th>Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Torsional stiff-ness</td>
<td>Load dependent adhesion</td>
<td>$k_{to,H} = \frac{dM_{to}}{d\phi} = \frac{8 \cdot G \cdot r_{K}^{3}}{3} \left[ 2 \left(1 - \frac{M_{to}}{M_{to,C,H}}\right)^{-1/2} \right]^{-1}$ (196)</td>
</tr>
<tr>
<td>Initial stiffness</td>
<td>Load dependent adhesion</td>
<td>$k_{to,H0} = \frac{dM_{to}}{d\phi}<em>{M</em>{to}=0} = 2 \cdot (1 - \nu) \cdot r_{1,2} \cdot (1 + \kappa) \cdot (F_{H0} + F_{N})$ (197)</td>
</tr>
<tr>
<td>Friction limits $k_{to,H} = 0$</td>
<td>Torsional moment</td>
<td>$M_{to,C,H} = \frac{2 \cdot \mu_{t}}{3} \left(1 + \kappa\right)^{3} \cdot (F_{N} + F_{H0}) \cdot \frac{3 \cdot \pi \cdot \kappa_{A} \cdot p_{f}}{4 \cdot G}$ (198)</td>
</tr>
<tr>
<td></td>
<td>Rotation angle</td>
<td>$\phi_{C,H} = \frac{3 \cdot \pi \cdot \mu_{t} \cdot \kappa_{A} \cdot p_{f}}{4 \cdot G}$ (200)</td>
</tr>
<tr>
<td>Loading $0 \leq \phi \leq \phi_{C,H}$</td>
<td>Moment - rotation angle</td>
<td>$M_{to} = 4 \cdot M_{to,C,H} \cdot \left[ \frac{3 \cdot \phi}{\phi_{C,H}^{3}} + 1 - \frac{3 \cdot \phi}{4 \cdot \phi_{C,H}} \right]^{-1}$ (199)</td>
</tr>
<tr>
<td>Unloading $-\phi_{C,H} \leq \phi \leq \phi_{C,H}$</td>
<td>El.-plastic, frictional</td>
<td>$M_{to} = M_{to,U} - 8 \cdot M_{to,C,H} \cdot \left[ \frac{3 \cdot (\phi_{U} - \phi)}{2 \cdot \phi_{C,H}} + 1 - \frac{3 \cdot (\phi_{U} - \phi)}{8 \cdot \phi_{C,H}} \right]^{-1}$ (202)</td>
</tr>
<tr>
<td>Reloading $-\phi_{C,H} \leq \phi \leq \phi_{C,H}$</td>
<td>El.-plastic, frictional</td>
<td>$M_{to} = -M_{to,U} + 8 \cdot M_{to,C,H} \cdot \left[ \frac{3 \cdot (\phi_{U} + \phi)}{2 \cdot \phi_{C,H}} + 1 - \frac{3 \cdot (\phi_{U} + \phi)}{8 \cdot \phi_{C,H}} \right]^{-1}$ (203)</td>
</tr>
</tbody>
</table>

4. Contact failure by shear and powder yield

The challenge is now how to apply these contact laws of history or load dependent adhesion forces $F_{H}(F_{N})$ to model the consolidation and flow of ultrafine cohesive powders, i.e. for applications in powder processing, storage and handling. Thus, the micro - macrotransition between particle contact and powder continuum behaviours should be accomplished here as simple as possible to obtain analytically the solutions of the powder yield limits which comprise the essential powder flow parameters [184, 185]. In computational micromechanics of granular matter, see e.g. [29 - 35, 38, 41 - 46, 236, 237], the more comprehensive volume averaged tensor notation is used to combine all the force vectors multiplied by position vectors distributed in a representative volume element (RVE) of particle packing under stress and strain.

Instead of this RVE, the model of a characteristic or representative particle contact (RPC) is used here to convert analytically the averaged normal and tangential contact forces into a two-dimensional stress state $\tau = f(\sigma)$. To formulate the failure conditions at representative particle contact of the powder shear zone, one can consider the Molerus’ theory [18]. But instead of the perfect plastic model [19], Eq. (72),

$$F_{H} = F_{H0} + \kappa_{p} \cdot F_{N}$$ (72)

the more complex, elastic-plastic particle contact model, Eq. (152), described in section 3.7.1,
\[
F_h = \frac{\kappa_A}{\kappa_A - \kappa_p} \cdot F_{h0} + \frac{\kappa_p}{\kappa_A - \kappa_p} \cdot F_N = (1 + \kappa) \cdot F_{h0} + \kappa \cdot F_N
\] (152)

is used to describe the failure conditions of the representative particle contact (RPC). Therefore following prerequisites and assumptions are necessary:

1. The contact areas are small in comparison to the characteristic particle size \(d\).
2. As a result of spherical symmetry, the contacts are uniformly distributed over the particle surface with the same probability.
3. The packing structure is assumed to be isotropic and random with uniform porosity \(\varepsilon\) in any cross-sectional areas as well as in the powder bulk.
4. Due to these assumptions an isotropic normal \(F_N\) and tangential force \(F_T\) distribution in the uniform distributed contacts is at least plausible. The average of these tangential forces is equivalent to the shear force resulting in the powder bulk \(\overline{F_T} = F_S\).
5. There is a direct correlation between isostatic stresses and contact forces as a superposition of three uniaxial normal stresses which are oriented orthogonal to each other.
6. Using the micro-microtransition of agglomerates derived first by Rumpf [76], the tensile strength of the powder packing (continuum) \(\sigma_Z\) can be correlated by porosity \(\varepsilon\), particle shape factor as surface ratio of volume-equivalent sphere and non-spherical particle \(\psi_A = A_{S,S_p} / A_{S,P}\), number function of particle contacts (expectation of coordination number) \(k(d)\), averaged sphere volume \(\nabla_p\) expressed by 3rd complete moment of cumulative particle size distribution \(M_{3,0}\)

\[
\nabla_p = \frac{\pi}{6} \cdot M_{3,0} = \frac{\pi}{6} \int d^3 \cdot q_0(d) \cdot d(d),
\] (205)

frequency distribution of particle size on number basis \(q_0(d)\) (index 0) and the adhesion force distribution \(F_H[d, q_0(d)]\) of all particle contacts in the failure zone:

\[
\sigma_Z = \frac{1 - \varepsilon}{6 \cdot \psi_A \cdot \nabla_p} \cdot \int_{d_{min}}^{d_{max}} k(d) \cdot F_H[d, q_0(d)] \cdot d(d) \cdot q_0(d) \cdot d(d) \approx \frac{1 - \varepsilon}{\varepsilon} \cdot \frac{F_{H}}{d^2}
\] (206)

7. Thus superposition and additivity in compression and shear mode provided, the general relation between stresses and contact forces for characteristic monodisperse particles of the equivalent size \(d \approx d_S\) (surface diameter) in a powder continuum amounts to [18]:

\[
\sigma, \tau = \frac{1 - \varepsilon}{\varepsilon} \cdot \frac{F_N, F_S}{d^2}
\] (207)

\[
\sigma_M, \sigma_R = \frac{1 - \varepsilon}{\varepsilon} \cdot \frac{F_M, F_R}{d^2}
\] (208)

8. With regard to the spherical symmetry and the uniform probability of contact normal forces a uniform angle between radius and centre contact force components \(F_{R \cdot \cos \alpha}, F_{M \cdot \cos \alpha}\) and normal vectors is designated by \(\alpha\).
Therefore, instead of radius \( \sigma_R \) and centre contact stresses \( \sigma_M \), the normal and shear force magnitudes orientated with this angle \( \alpha \) against direction of major principal stress may be generally expressed by:

\[
F_N = F_M + F_R \cdot \cos 2\alpha \tag{209}
\]

\[
F_S \leq F_R \cdot \sin 2\alpha \tag{210}
\]

If the shear force should be completely activated in the shear zone, then the ‘=’ is valid in the last Eq. (210).

With regard to the uniform probability of contact forces and stresses, uniform coefficients of internal friction at contacts \( \mu_i \) and angles of internal friction \( \phi_i \) in the bulk powder are assumed, i.e. \( \mu_i = \tan \phi_i \approx \text{const.} \)

These prerequisites are necessary to obtain comfortable algebraic constitutive equations for the stress limits of cohesive powder yield. By this restricted method, the force - displacement behaviour of the adhesion contacts, which are in a scale of plastic strain \( h_K/d \approx 10^{-4} \) according to Fig. 10, could not be directly transferred into the stress - strain behaviour of the powder having a plastic strain of about \( \Delta h/h_{Sz} \approx 0.05 \). Schubert [82, 83] has discussed this still existing problem by the elastic-plastic tensile stress - strain behaviour of moist agglomerates. Generally, this micro - macrotransition can only be accomplished by sophisticated numerical models implemented in the discrete element method (DEM) [157, 158, 194, 195, 327, 338, 339]. But despite this restriction, the contact force models of representative particle contact should be used here to obtain the stress relations of the powder yield limits which can comfortably and analytically be handled.

According to the condition of Coulomb friction Eq. (174), the ratio of shear force/sum of normal forces is used to describe simultaneous sliding or steady-state flow at particle contacts under compression:

\[
\frac{F_S}{\sum F_{N,i}} = \frac{F_R \cdot \sin 2\alpha}{(1 + \kappa) \cdot F_{h0} + (1 + \kappa) \cdot (F_M + F_R \cdot \cos 2\alpha)} \leq \frac{\tan \phi_i}{\tan \phi_{st}} \tag{211}
\]

To eliminate the failure angle \( \alpha \) from Eq. (211), equality is only achieved for a value of \( \alpha \) where the left-hand side in brackets is a maximum [18]. This is valid for the common relation of the characteristic orientation of normal of the shear zone against direction of major principal stress:

\[
\alpha = \frac{\pi}{4} + \frac{\phi_{st}}{2} \tag{212}
\]

The angle of internal friction \( \phi_i \) for incipient contact failure (slope of linear yield locus) and the stationary angle of internal friction \( \phi_{st} \) can be combined as follows [18, 155]:

\[
\tan \phi_{st} = (1 + \kappa) \cdot \tan \phi_i \tag{213}
\]

The softer the particle contacts, the larger are the differences between these friction angles and consequently, the more cohesive is the powder response.
The condition for incipient or instantaneous contact failure under compression is consequently as ratio of sum of shear forces/sum of normal forces ($F_{HM}$ and $F_{HR}$ are the adhesion force components of contact preconsolidation)

$$\sum F_{S,j} = F_R \cdot \left[ \sin 2\alpha - \left( 1 + \kappa \cdot F_{HR} / F_R \right) \cdot \tan \varphi_i \cdot \cos 2\alpha \right]$$

but now with an auxiliary angle $\beta$:

$$\tan \beta = \left( 1 + \kappa \cdot F_{HR} / F_R \right) \cdot \tan \varphi_i$$

Equality in Eq. (214) is only achieved for a value of $\alpha$ where the left-hand side in brackets is a maximum. This is valid for the failure angle $\alpha$ of the characteristic orientation of contact forces within the shear zone [18]:

$$\alpha = \frac{\pi}{4} + \frac{\beta}{2}$$

The larger the term of Eq. (215) in brackets $\kappa \cdot \sigma_{VR} / \sigma_R \geq 0$, expressed here as the ratio of radius stresses for consolidation pre-history $\sigma_{VR}$ and yielding $\sigma_R$, the more is the difference from the normally expected relation of $\alpha = \pi / 4 + \varphi_i / 2$. This latter relation is predicted to be valid only for stiff particle contacts without any contact consolidation $\kappa = 0$.

Next, the auxiliary angle $\beta$ has to be eliminated from Eq. (214) and a square function is obtained with the positive root:

$$F_R = \sin \varphi_i \left[ \sqrt{\left( (1 + \kappa) \cdot F_{H0} + \kappa \cdot F_{HM} + F_M \right)^2 - \kappa^2 \cdot F_{HR}^2 \cdot \cos^2 \varphi \cdot \varphi_i - \kappa \cdot F_{HR} \cdot \sin \varphi \cdot \varphi_i} \right]$$

This model is expressed in stresses, i.e. from Eq. (208) the preconsolidation forces $F_{HR}, F_{HM}$ are transferred into radius $\sigma_{VR}$ and centre stresses $\sigma_{VM}$ of the compression states. By Eqs. (217) and (213) the general non-linear equation of the yield locus of a cohesive powder is obtained:

$$\sigma_R = \sin \varphi_i \cdot \sigma_{VR} \cdot \left[ \left( \frac{\tan \varphi_i \cdot (\sigma_{VM} + \sigma_0) - \sigma_{VM} + \sigma_M}{\sigma_{VR}} \right)^2 - \frac{\sin^2 (\varphi_i - \varphi)}{\cos^2 \varphi_i \cdot \tan^2 \varphi_i} - \frac{\sin (\varphi_i - \varphi)}{\cos \varphi_i} \right]$$

This constitutive equation (218) combines the radius $\sigma_R$ and centre $\sigma_M$ of Mohr circles of incipient yield with radius $\sigma_{VR}$ and centre $\sigma_{VM}$ of Mohr circles of arbitrary preconsolidation state as envelope. This instantaneous yield locus describes the limit of incipient plastic deformation during powder yield, Fig. 22. The elastic domain of the cohesive powder is located below this yield limit and depends on the characteristic preconsolidation stresses [185].

It is worth to note here that the stressing pre-history of cohesive powder flow is stationary (steady-state) and, from Eqs. (208) and (211), results significantly in a cohesive stationary yield locus. This stationary yield locus is expressed in radius stress-centre stress coordinates (radius $\sigma_{VR} = \sigma_{R, st}$ and centre $\sigma_{VM} = \sigma_{M, st}$ of Mohr circle for steady-state flow):

$$\sigma_{R, st} = \sin \varphi_i \cdot (\sigma_{M, st} + \sigma_0)$$
This steady-state powder flow is characterised by a dynamic equilibrium of simultaneous contact shearing, unloading and failing, creating new contacts, loading, reloading, unloading and shearing again within a shear zone [338]. Thus, the stationary yield locus is the envelope of all Mohr circles for steady-state flow with a certain intersection of the abscissa $\sigma_0$ as isostatic tensile strength of an unconsolidated powder:

$$\sigma_0 = \frac{1 - e_0}{e_0} \cdot \frac{F_{H0}}{d^2}$$  \hspace{1cm} (220)

$F_{H0}$ characterises an average adhesion force without any contact deformation of an adhesion force distribution within the powder and $\varepsilon_0$ is the porosity of the loose powder packing.

$$\sigma = \sin \varphi_i \cdot (\sigma_{M,\text{st}} + \sigma_0) \cdot \frac{\tan \varphi_{\text{st}}}{\tan \varphi_i} \cdot \left[ 1 - \tan \varphi_i \cdot \frac{\sigma_{M,\text{st}} - \sigma_{M}}{\sigma_{M,\text{st}} + \sigma_0} \right] - \sin^2(\varphi_{\text{st}} - \varphi_i) - \tan \varphi_i \cdot \sin(\varphi_{\text{st}} - \varphi_i)$$

$$\sigma_{M,\text{st}}$$

Substitution of Eq. (219) in Eq. (218) results in the general non-linear equation of the yield locus of a cohesive powder with steady-state flow as preconsolidation history:

$$\sigma = \sin \varphi_i \cdot (\sigma_{M,\text{st}} + \sigma_0) \cdot \frac{\tan \varphi_{\text{st}}}{\tan \varphi_i} \cdot \left[ 1 - \tan \varphi_i \cdot \frac{\sigma_{M,\text{st}} - \sigma_{M}}{\sigma_{M,\text{st}} + \sigma_0} \right] - \sin^2(\varphi_{\text{st}} - \varphi_i) - \tan \varphi_i \cdot \sin(\varphi_{\text{st}} - \varphi_i)$$

$$\sigma_{M,\text{st}}$$

The general non-linear yield locus of a cohesive powder Eq. (221) is completely described only with three material parameters plus the influence of preconsolidation stress $\sigma_{M,\text{st}}$:

1. $\varphi_i$ – incipient particle friction of failing contacts, i.e. Coulomb friction;
(2) \( \varphi_{st} \) – steady-state particle friction of failing contacts, increasing adhesion by means of flattening of contact expressed with the contact consolidation coefficient \( \kappa \), or by friction angles \((\varphi_{st} - \varphi_t)\) as shown in Eq. (218). The softer the particle contacts, the larger is 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 adhesion force in an unconsolidated powder;

(4) \( \sigma_{M,st} \) – previous consolidation influence of an additional normal force at particle contact, characteristic centre stress of Mohr circle of preconsolidation 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.

With this as the physical basis, a complete set of models for incipient powder consolidation, yield and steady-state flow was derived. The yield surface due to theory of plasticity was expressed by simple linear equations in centre and radius stresses [180, 184, 185, 271]. The preconsolidation stress \( \sigma_{M,st} \) is also the key parameter of the so-called compression functions, i.e. the compression rate \( \frac{d \rho_b}{d \sigma_{M,st}} = f(\sigma_{M,st}) \), bulk density \( \rho_b = f(\sigma_{M,st}) \) and the mass-related compression work \( W_{m,b} = f(\sigma_{M,st}) \), the specific power consumption \( P_{m,b,pre} = f(\sigma_{M,st}) \) of steady-state flow and the mass-related preshear work \( W_{m,b,pre} = f(\sigma_{M,st}) \) of the cohesive powder which were explained in a previous paper [185].

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 [338, 339].

5. Conclusions

The models for elastic (Hertz, Huber, Fromm, Sonntag, Mindlin & Deresiewicz), elastic-adhesion (Derjaguin, Johnson, Thornton), plastic-adhesion (Derjaguin, Krupp, Molerus, Johnson, Maugis and Pollock) contact deformation response of a single, normal and tangential loaded, isotropic, smooth contact of two spheres were discussed. The force-displacement behaviours of elastic-plastic (Schubert, Thornton), elastic-dissipative (Sadd), plastic-dissipative (Walton & Braun), viscous (Cundall, Pöschel, Stieß) and viscoplastic-adhesive (Rumpf) contacts were also shown. With respect to these theories, a general approach for the time and deformation rate dependent and combined viscoelastic, plastic, viscoplastic, adhesion and dissipative behaviours of a contact between spherical particles was derived and explained.

As the main result, the total adhesion force \( F_{H}(F_N) \) is found to be a function of the force contribution \( F_{H0} \) without any deformation plus a consolidation pre-history or load dependent term with the normal force \( F_N \). These linear and non-linear approaches can be interpreted as general constitutive models of the adhesion force. It should be pointed out here that the adhesion force level discussed in this paper is approximately \( 10^4 \) - \( 10^6 \) times the particle weight of ultrafine particles. This means, in other words, that one has to apply these large values as acceleration ratio \( a/g \) with
respect to gravity to separate these pre-consolidated contacts or to remove mechanically such adhered particles from solid surfaces.

In terms of *frictional shear flow* of cohesive ultrafine powders, the essential contribution of pre-consolidation or, in other words, load dependent adhesion \( F_H(F_N) \) on the tangential force in an elastic-plastic frictional contact is demonstrated. Rolling resistance and elastic-plastic frictional twisting with load dependent adhesion are calculated as well. The energy dissipation by friction work due to contact shear, unload and reverse shear, is very large compared to the specific energy absorption by normal load, unload and reload.

The coefficient of restitution and maximum impact force response are also derived for adhesive elastic-plastic contact behaviours in terms of *rapid collisional shear flow* of cohesive ultrafine powders. For colliding adhesive particles a correlation between particle impact velocity and contact displacement response is obtained using energy balance.

These constitutive model approaches are generally applicable for micro- and nanocontacts of particulate solids [155, 178, 271]. For a representative particle contact (RPC), contact force models are used to obtain the stress relations of the yield limits. Hence, these contact models are intended to be applied for modern data evaluation of product quality characteristics such as powder flow properties, i.e., yield loci, consolidation and compression functions or design of characteristic processing apparatus dimensions [98, 152, 179, 184, 185, 329 - 340].

**Acknowledgements**


The advices from H.-J. Butt, L. Heim, M. Kappl and S. Luding with respect to the fundamentals of particle and powder mechanics were especially appreciated during the collaboration of the project “shear dynamics of cohesive, fine-disperse particle systems“ of the joint research program “Behaviour of Granular Media“ of German Research Association (DFG).

**Symbol, Unit, Description**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>nm</td>
<td>contact separation</td>
</tr>
<tr>
<td>A</td>
<td>mm^2</td>
<td>cross-sectional area</td>
</tr>
<tr>
<td>( A_K )</td>
<td>nm^2</td>
<td>particle contact area</td>
</tr>
<tr>
<td>( a_{F=0} )</td>
<td>nm</td>
<td>minimum centre separation for force equilibrium of molecular attraction and repulsion potentials</td>
</tr>
<tr>
<td>( A_{S,m} )</td>
<td>m^2/g</td>
<td>mass related surface area</td>
</tr>
<tr>
<td>b</td>
<td>Ns/m</td>
<td>damping coefficient</td>
</tr>
<tr>
<td>( b_K )</td>
<td>nm</td>
<td>contact width</td>
</tr>
<tr>
<td>c</td>
<td>Ns/m</td>
<td>contact damping coefficient</td>
</tr>
<tr>
<td>Ca</td>
<td>-</td>
<td>capillary number</td>
</tr>
<tr>
<td>( C_H )</td>
<td>J</td>
<td>Hamaker constant [210] based on interacting molecule pair additivity</td>
</tr>
<tr>
<td>Symbol</td>
<td>Unit</td>
<td>Definition</td>
</tr>
<tr>
<td>--------</td>
<td>------</td>
<td>------------</td>
</tr>
<tr>
<td>$C_{H,ls}$</td>
<td>J</td>
<td>Hamaker constant according to Lifshitz theory [211] for solid-liquid-solid interaction</td>
</tr>
<tr>
<td>d</td>
<td>µm</td>
<td>particle size</td>
</tr>
<tr>
<td>D</td>
<td>-</td>
<td>damage (fractional reduction in stiffness)</td>
</tr>
<tr>
<td>e</td>
<td>-</td>
<td>coefficient of restitution</td>
</tr>
<tr>
<td>E</td>
<td>kN/mm$^2$</td>
<td>modulus of elasticity</td>
</tr>
<tr>
<td>F</td>
<td>N</td>
<td>force</td>
</tr>
<tr>
<td>$F_{H}$</td>
<td>nN</td>
<td>adhesion force</td>
</tr>
<tr>
<td>$F_{H0}$</td>
<td>nN</td>
<td>adhesion force of a rigid contact without any contact deformation, see Eq. (158)</td>
</tr>
<tr>
<td>$F_N$</td>
<td>nN</td>
<td>normal force</td>
</tr>
<tr>
<td>$F_S$</td>
<td>nN</td>
<td>shear force of characteristic contacts of bulk powder</td>
</tr>
<tr>
<td>$F_T$</td>
<td>nN</td>
<td>tangential force of single contact</td>
</tr>
<tr>
<td>G</td>
<td>kN/mm$^2$</td>
<td>shear modulus</td>
</tr>
<tr>
<td>h</td>
<td>mm</td>
<td>zone height</td>
</tr>
<tr>
<td>$h_K$</td>
<td>nm</td>
<td>height of overlap, indentation or centre approach</td>
</tr>
<tr>
<td>$h_r$</td>
<td>nm</td>
<td>radius of a spherical asperity on particle surface</td>
</tr>
<tr>
<td>i</td>
<td>-</td>
<td>number of fractions</td>
</tr>
<tr>
<td>k</td>
<td>-</td>
<td>coordination number</td>
</tr>
<tr>
<td>$k_N$</td>
<td>N/mm$^2$</td>
<td>contact stiffness in normal direction</td>
</tr>
<tr>
<td>$k_T$</td>
<td>N/mm$^2$</td>
<td>contact stiffness in tangential direction</td>
</tr>
<tr>
<td>l</td>
<td>nm</td>
<td>particle length</td>
</tr>
<tr>
<td>m</td>
<td>g</td>
<td>mass</td>
</tr>
<tr>
<td>M</td>
<td>Nm</td>
<td>moment (torque)</td>
</tr>
<tr>
<td>$M_{k,r}$</td>
<td>µm$^r$</td>
<td>k-th statistical moment of particle size distribution on quantity basis r</td>
</tr>
<tr>
<td>n</td>
<td>-</td>
<td>number of stressing events</td>
</tr>
<tr>
<td>p</td>
<td>kPa</td>
<td>contact pressure</td>
</tr>
<tr>
<td>$p_f$</td>
<td>MPa</td>
<td>plastic micro-yield strength of particle contact</td>
</tr>
<tr>
<td>$p_m$</td>
<td>MPa</td>
<td>average contact pressure at confined elastic-plastic yield</td>
</tr>
<tr>
<td>$p_{VdW}$</td>
<td>MPa</td>
<td>attractive van der Waals pressure, see Eq. (40)</td>
</tr>
<tr>
<td>q(d)</td>
<td>µm$^{-1}$</td>
<td>quantity of particle size frequency distribution</td>
</tr>
<tr>
<td>Q(d)</td>
<td>-</td>
<td>quantity of cumulative distribution</td>
</tr>
<tr>
<td>r</td>
<td>µm</td>
<td>particle radius</td>
</tr>
<tr>
<td>$r_K$</td>
<td>nm</td>
<td>contact radius</td>
</tr>
<tr>
<td>s</td>
<td>µm</td>
<td>displacement, macroscopic shear displacement</td>
</tr>
<tr>
<td>$s_B$</td>
<td>µm</td>
<td>critical displacement at particle breakage</td>
</tr>
<tr>
<td>$S_N$</td>
<td>m$^{-1}$</td>
<td>slope of unload stiffness according to Walton [145]</td>
</tr>
<tr>
<td>t</td>
<td>s</td>
<td>time</td>
</tr>
<tr>
<td>$T_K$</td>
<td>ns</td>
<td>contact duration at elastic impact</td>
</tr>
<tr>
<td>v</td>
<td>m/s</td>
<td>particle velocity</td>
</tr>
<tr>
<td>V</td>
<td>m$^3$</td>
<td>volume</td>
</tr>
<tr>
<td>$v_{IH}$</td>
<td>m/s</td>
<td>critical sticking velocity</td>
</tr>
<tr>
<td>$v_{ij}$</td>
<td>m/s</td>
<td>compression rates of strain tensor</td>
</tr>
<tr>
<td>$v_{ij}$</td>
<td>m/s</td>
<td>shear rates of strain tensor</td>
</tr>
<tr>
<td>$v_R$</td>
<td>m/s</td>
<td>bounce velocity</td>
</tr>
<tr>
<td>$v_S$</td>
<td>m/s</td>
<td>shear rate</td>
</tr>
<tr>
<td>$v_{\phi}$</td>
<td>m/s</td>
<td>circumferential speed</td>
</tr>
<tr>
<td>W</td>
<td>J</td>
<td>deformation work</td>
</tr>
<tr>
<td>$W_m$</td>
<td>J/g</td>
<td>mass related energy absorption by inelastic deformation</td>
</tr>
<tr>
<td>x, y, z</td>
<td>mm</td>
<td>spatial coordinates of volume element</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>deg</td>
<td>failure angle of shear plane</td>
</tr>
<tr>
<td>$\alpha_{R,slip}$</td>
<td>-</td>
<td>micro-slip coefficient of rolling resistance</td>
</tr>
</tbody>
</table>
\( \beta \)  \( \text{deg} \)  auxiliary angle
\( \beta_d \)  \( s \)  local damping factor according to Cundall [41]
\( \gamma \)  \( \text{deg} \)  rolling angle, distortion
\( \gamma_d \)  -  damage accumulation index
\( \delta \)  \( \text{nm} \)  tangential contact displacement
\( \varepsilon \)  -  porosity
\( \varepsilon_{ii} \)  -  normal strains of strain tensor
\( \varepsilon_{ij} \)  -  distortions of strain tensor
\( \eta_p \)  Pa\cdot s  apparent particle (solid) viscosity
\( \eta_K \)  Pa\cdot s  apparent contact viscosity
\( \Theta \)  -  plastic yield factor at indentation according to Stronge [55]
\( \Theta \)  -  unit step function or Heaviside function: \( \Theta(x \leq 0) = 0 \) and \( \Theta(x > 0) = 1 \)
\( \kappa \)  -  elastic-plastic contact consolidation coefficient, see Eq. (151)
\( \kappa_A \)  -  elastic-plastic contact area coefficient, see Eq. (95)
\( \kappa_p \)  -  plastic repulsion coefficient, see Eq. (73)
\( \kappa_{pa} \)  -  viscoplastic repulsion coefficient, see Eq. (101)
\( \kappa_{vis} \)  -  total viscoplastic contact consolidation coefficient, see Eq. (157)
\( \mu_A \)  kg/kg  mass fraction of flow additive in a cohesive powder
\( \mu_i \)  -  coefficient of internal particle-particle friction, i.e. Coulomb friction
\( \nu \)  -  Poisson ratio
\( \rho \)  kg/m\(^3\)  density
\( \rho_b \)  kg/m\(^3\)  bulk density of powder
\( \sigma \)  kPa  normal stress
\( \sigma_{ii} \)  kPa  normal stresses of stress tensor
\( \sigma_{ij} \)  kPa  shear stresses of stress tensor
\( \sigma_M \)  kPa  centre stress of Mohr circle [2, 271]
\( \sigma_R \)  kPa  radius stress of Mohr circle [2, 271]
\( \sigma_{sls} \)  mJ/m\(^2\)  surface tension of solid-liquid-solid interaction
\( \sigma_t \)  kPa  tensile stress
\( \sigma_0 \)  kPa  isostatic tensile strength of the unconsolidated powder
\( \sigma_1 \)  kPa  major principal stress
\( \sigma_2 \)  kPa  minor principal stress
\( \tau \)  kPa  shear stress
\( \phi \)  -  rotation angle
\( \varphi_i \)  deg  angle of internal friction between particles
\( \varphi_{at} \)  deg  stationary angle of internal friction
\( \Phi_T \)  -  dimensionless bond strength according to Tabor [208]
\( \psi \)  -  loading parameter according to Thornton [131]
\( \psi_A \)  -  particle shape factor (spherericity) according to Wadell [325]
\( \omega \)  s\(^{-1}\)  angular velocity
\( \omega_0 \)  s\(^{-1}\)  eigenfrequency of a particle

Indices

A  detachment or contact area related
at  attraction
b  bulk
br  brittle
c  compressive
C Coulomb, cantilever
crit critical
diss dissipation
e effective
el elastic
f flow or yield
F=0 potential force equilibrium (potential minimum)
gl global
H adhesion
i internal, spatial coordinates
iso isostatic
j number index, spatial coordinates
K particle contact
l liquid
m mass related, average
M centre
min minimum
n number index
N normal
p pressure related
P particle
pl plastic
r micro-roughness
R rolling resistance, radius
rep repulsion
rot rotation
s solid
S surface, shear
sls solid-liquid-solid interaction between particle surfaces
sp sphere
ss solid-vacuum-solid interaction between particle surfaces
st stationary
Sz shear zone
t loading time dependent
to torsion
T tangential
th theoretical
tot total
U unloading
V volume related
VdW van der Waals
vis total viscoplastic
x, y, z spatial coordinate directions
0 initial, zero point, number basis of cumulative distribution
(0) beginning of iterations
1,2 particle 1, particle 2
3 mass basis of cumulative distribution of particle size (d³)
50 median particle size, i.e. 50 % of cumulative distribution
63 characteristic argument x/x₆₃ = 1 of any exponential function at which 1-exp(-x/x₆₃) = 0.63
References

[34] J.D. Goddard, p. 129.133, in
[89] A. Saito, and K. Ota, in [87] pp. 105-114
[113] B.V. Derjaguin, Kolloid Zeitschr. 69, 155-164 (1934)
[115] L. Föppl, Die strenge Lösung für die rollende Reibung, Leibnitz Verlag, München (1947)
[119] L. Föppl, Die strenge Lösung für die rollende Reibung, Leibnitz Verlag, München (1947)
[125] N. Maw, J.R. Barber and J.N. Fawcett, Wear 38, 101-114 (1976)
[209] H.C. Hamaker, Physica 4, 1058-1072 (1937)
[286] D. Sarid, Scanning force microscopy: with applications to electric, magnetic and atomic forces, Oxford University Press (1991)
(2006) manuscript