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Pressure-dependent bulk compressibility of a porous granular material modelled by improved contact mechanics and micromechanical approaches: effects of surface roughness of grains

Lei Wang^{a,b,*}, Georg Dresen^{a,c}, Erik Rybacki^a, Audrey Bonnelye^a, Marco Bohnhoff^{a,b}

^a Helmholtz Centre Potsdam, GFZ German Research Centre for Geosciences, 14473 Potsdam, Germany

^b Department of Earth Sciences, Free University Berlin, 12249 Berlin, Germany

^c Institute of Earth and Environmental Science, University of Potsdam, 14469 Potsdam, Germany

*Corresponding author

Tel.: +49 331 288-28835, Email address: lei.wang@gfz-potsdam.de (Lei Wang)

Abstract

The change of the mechanical properties of granular materials with pressure is an important topic associated with many industrial applications. In this paper we investigate the influence of hydrostatic pressure (P_e) on the effective bulk compressibility (C_{eff}) of a granular material by applying two modified theoretical approaches that are based on contact mechanics and micromechanics, respectively. For a granular material composed of rough grains, an extended contact model is developed to elucidate the effect of roughness of grain surfaces on bulk compressibility. At relatively low pressures, the model predicts that the decrease of bulk compressibility with pressure may be described by a power law with an exponent of $-1/2$ (i.e., $C_{eff} \propto P_e^{-1/2}$), but deviates at intermediate pressures. At elevated pressures beyond full contact, bulk compressibility remains almost unchanged, which may be roughly evaluated by continuum contact mechanics. As an alternative explanation of pressure-dependent bulk compressibility, we suggest a micromechanical model that accounts for effects of different types of pore space present in granular materials. Narrow and compliant inter-granular cracks are approximated by three-dimensional oblate spheroidal cracks with rough surfaces, whereas the equant and stiff pores surrounded by three and four neighbouring grains are modelled as tubular pores with cross sections of three and four cusp-like corners, respectively. In this model, bulk compressibility is strongly reduced with increasing pressure by progressive closure of rough-walled cracks. At pressures exceeding crack closure pressure, deformation of the remaining equant pores is largely insensitive to pressure, with almost no further change in bulk compressibility. To validate these models, we performed hydrostatic compression tests on Bentheim sandstone (a granular rock consisting of quartz with high porosity) under a wide range of pressure. The relation between observed microstructures and measured pressure-dependent bulk compressibility is well explained by both suggested models.

Keywords: bulk compressibility; granular materials; roughness; micromechanical model; contact model

1. Introduction

Evaluating the mechanical properties of porous granular materials is a topic of high interest due to its industrial applications, ranging from oil and gas exploitation from porous sandstone reservoirs to pharmaceutical fabrication processes. In contrast to other solid materials like glass and crystalline metal, the elastic properties of porous granular materials generally increase nonlinearly with isotropic pressure [1]. Such a nonlinear elastic response of granular media is, in principle, intrinsically related to their microstructural characteristics [2,3]. It is commonly attributed either to the non-linear change of the contact area between two elastic grains (i.e., contact mechanics approach) [4], or to the existence of a network of microcracks in the solid phase (i.e., micromechanical approach) [5-7]. Both approaches are briefly summarized in the following.

First, we consider predictions on the evolution of bulk compressibility with increasing pressure based on contact theory. According to Hertz contact theory, the contact law for grain-to-grain interaction [8] under the framework of continuum contact mechanics predicts that the bulk compressibility (C_{eff}) of a granular aggregate scales with effective hydrostatic pressure (P_e) following a power law relationship with an exponent of $-1/3$, (i.e., $C_{eff} \propto P_e^{-1/3}$).

This relation arises from the non-linear increase in contact area between two smooth spheres with increasing hydrostatic pressure. Experimental results, however, indicate that for a random packing of sand grains or spherical glass beads, the exponent typically varies between $-1/3$ and $-1/2$ [1,4,9]. This deviation from Hertz theory prediction is explained by the dependence of the coordination number (the average contact number per particle) on pressure or on porosity [9-11]. Hertz theory is found to be more appropriate for unconsolidated granular materials with high porosity (36 % – 40 %) close to suspensions. In this case, the material may be geometrically described by a dense random packing of elastic spheres with a coordination number of about 9 [12-14]. For the granular material with a porosity of less than 36%, the properties and positions of intergranular bonds at grain contacts strongly influence the macroscopic mechanical behaviour. For example, an increase in contact area between two grains by cementation or consolidation reduces porosity and increases the contact stiffness [13,15-19]. All contact models aforementioned rely on the common assumption that all grains are absolutely smooth. However, general particles in granular aggregate such as sandstones, concretes, or packed powders usually have rough surfaces. The contact area is restricted to isolated asperities that sustain the normal force acting on them under pressure, which leads at relatively low pressures to an overestimation of the contact stiffness between grains by using continuum contact mechanics (e.g., Hertz-based contact models for smooth surfaces) [20].

Two classes of analytical contact models were developed to investigate the pressure-dependent contact area between two elastic solid bodies with nominally flat but microscopically rough surfaces. The first category is fundamentally derived from a statistical description of the topography of rough surfaces containing asperities, here classified as asperity-based models [21-39]. In these models, the Hertzian contact solution for the contact between two smooth spheres is adopted to characterize the mechanical behaviour of a single asperity, the tip of which is mostly assumed to be spherical or elliptical with constant or varying curvature. The macroscopic contact behaviour between two blocks is modelled as a sum of a statistical contribution of non-interacting and contacting asperities. In contrast, an alternative contact model proposed by Persson [40,41] considers the relation between randomly rough surface heights and associated heterogeneous contact pressure distributions in

the limit of full contact. Persson's model gives the exact solution for full contact conditions by deriving a diffusion equation for the probability density of the contact pressure that depends on the variance of the surface roughness. Interestingly, both types of models predict a nearly linear relation between real contact area and normal force acting on contact surface in the limit of infinitesimally small contact. However, so far these models have been only found to successfully model two solid blocks contacting along rough surfaces [42-50], but not for the case of a granular material.

Theoretical approaches based on micromechanics argue that at low pressures the non-linearity of compressibility with pressure in porous media arises from the existence of a network of soft voids (microcracks) in the solid phase [5,6,51,52]. The shape of voids in granular materials are highly variable, but, to first order, the total pore space (total porosity) may be divided into stiff, low-compliant (pore) porosity and compliant (crack) porosity [53,54], which form an interconnected pore network. Although the crack porosity is often very low compared to stiff porosity, it plays a significant role in affecting the mechanical properties of granular materials. A compliant crack is commonly represented by a strongly oblate spheroidal void [55-60]. The closure of such a crack under pressure depends on its aspect ratio, which is the ratio of width to length of the spheroid [53,61]. The stiff pores are mostly simplified as either two-dimensional circles or three-dimensional spheres. The compressibility of stiff pores usually shows a linear relation with porosity [3,62,63].

Assuming that a combination of compliant cracks and stiff pores are randomly embedded into a solid matrix, a diversity of micromechanical models was developed to estimate the effective macroscopic elastic properties of porous materials [5,6,53,55-58,64-66]. In these micromechanical models, the effect of cracks on the elastic properties is dominated by the crack density tensor, which is frequently used to interpret seismic or ultrasonic wave data. Unfortunately, the crack density is generally not directly measured in hydrostatic compression tests. Instead, in many cases it is derived from measurements of the static or dynamic elastic moduli. Moreover, representing the cracks by simplified smooth oblate spheroidal voids exaggerates their compliant behaviour with respect to that of real cracks in granular materials [60]. In contrast, approximating the stiff pores by spheres or circles generally overestimates the stiffness of granular materials. At least within general granular materials, the complex pore structure is hardly represented by such simple geometrical approximations.

The objective of this paper is to probe the pressure-dependent bulk compressibility of porous granular materials based on improved analytical models that rely on contact mechanics and micromechanics taking into account the roughness of grain surfaces. To test the applicability of the models on the mechanical behaviour of porous granular materials, we conducted hydrostatic compression tests on Bentheim sandstone (a granular rock consisting of quartz with high porosity) under a wide range of pressure. The relation between observed microstructures and measured pressure-dependent bulk compressibility is demonstrated by both suggested models.

2. Bulk compressibility of a rough granular material: an extended contact mechanics approach

Typically, continuum contact mechanics for smooth surfaces relates the effective bulk compressibility C_{eff} of granular materials to the real contact area A between identical spherical grains [8,15], as expressed by:

$$C_{eff} = \frac{3\pi(1-\nu_m)}{Z(1-\phi)G_m} \left(\frac{A}{\pi R^2} \right)^{-1/2} \quad (1)$$

where ν_m and G_m are the Poisson's ratio and shear modulus of grains, respectively, ϕ is porosity, Z is the coordination number related to porosity or pressure, and R is the average radius of grains.

Let us now consider the case of a granular material composed of nominally spherical particles with microscopically rough surfaces, as illustrated in Fig. 1. Assuming that the initial apparent contact area between two grains is A_0 , the real contact area A should be lower than A_0 .

In the case of infinitesimally small contact between semi-infinite solids with rough surfaces at low pressures, both asperity-based models and Persson's model predict a linear growth of the real contact area with pressure, as formulated by [31,39,42-44,67]:

$$\frac{A}{A_0} = \frac{\kappa}{\sqrt{\langle |\nabla h|^2 \rangle}} \frac{p_0}{E'} \quad (2)$$

where $\sqrt{\langle |\nabla h|^2 \rangle}$ is the root-mean-square (rms) gradient of surface topography where $h(\mathbf{x})$ is height as a function of position $\mathbf{x}$ in the contact plane, and p_0 is the nominal normal stress defined as $p_0 = F/A_0$ in which F is the applied normal force between two rough surfaces. E' is the effective Young's modulus given by $1/E' = (1-\nu_1^2)/E_1 + (1-\nu_2^2)/E_2$ where ν_i and E_i are the Poisson ratios and Young's moduli of the two solid blocks if they have different mechanical properties. The proportionality coefficient κ is found to be equal $\sqrt{2\pi}$ in asperity-based models and $\sqrt{8/\pi}$ in Persson's model for the asymptotic limit at infinitesimally low pressure [42,43].

Provided that the curvature radius of asperities is negligible compared to the radius of the whole grain and that the little change of A_0 between two rough grains with pressure occurs (for large A_0 at non-stressed state), Eq. (2) may then be extended to represent change of contact area for a rough grain assembly.

The induced normal force F acting on identical spheres due to hydrostatic compression in the Hertz-based contact solution is given by [8,15]:

$$F = \frac{4\pi R^2 P_e}{Z(1-\phi)} \quad (3)$$

where P_e is the effective hydrostatic pressure (difference between confining pressure P_c and inner pore pressure P_p). Substituting Eq. (3) into Eq. (2), the real contact area between rough grains normalized to their cross-section area ($A/\pi R^2$) is given by:

$$\frac{A}{\pi R^2} = \frac{\kappa}{\sqrt{\langle |\nabla h|^2 \rangle}} \frac{4P_e}{ZE'(1-\phi)} \quad (4)$$

Substituting Eq. (4) into Eq. (1) yields the pressure-dependent bulk compressibility of granular media at low pressures:

$$C_{eff} = \left[\frac{4ZG_m^2(1-\phi)P_e}{9\pi^2 E'(1-\nu_m)^2} \frac{\kappa}{\sqrt{\langle |\nabla h|^2 \rangle}} \right]^{-1/2} \quad (5)$$

Eq. (5) suggests that the effective bulk compressibility of granular material at low pressures depends on $\sqrt{\langle |\nabla h|^2 \rangle}$ (i.e., surface roughness) in a way that the rougher surfaces of grains yield higher bulk compressibility. Assuming that Z and ϕ change little under pressure, C_{eff} is proportional to $P_e^{-1/2}$ at low pressures, contrary to $P_e^{-1/3}$ for Hertz contact model (see Appendix B1 for Hertz model in detail). Note that Eq. (5) is valid when grain surfaces are rough. Towards the limit of full contact along the grain interface at high pressures, Eq. (5) may become increasingly inaccurate since Eq. (2) does not consider possible interaction and coalescence between adjacent growing contact zones [42,68,69].

In Persson's model, the evolution of the real contact area fraction with pressure over the whole range from infinitesimal contact to full contact can be described by an error function [40,41]:

$$\frac{A}{A_0} = \text{erf} \left(\frac{p_0}{E'} \sqrt{\frac{2}{\langle |\nabla h|^2 \rangle}} \right) \quad (6)$$

Note that Eq. (2) is the asymptotic form of Eq. (6) at infinitesimally low pressure for $\kappa = \sqrt{8/\pi}$. Combining Eq. (3) with Eq. (6), the real contact area normalized by the grain cross-section area ($A/\pi R^2$) may then be estimated by:

$$\frac{A}{\pi R^2} = \frac{A_0}{\pi R^2} \text{erf} \left(\frac{\pi R^2}{A_0} \frac{4P_e}{ZE'(1-\phi)} \sqrt{\frac{2}{\langle |\nabla h|^2 \rangle}} \right) \quad (7)$$

Substituting Eq. (7) into Eq. (1), the evolution of bulk compressibility of granular materials over a wide range of pressure is given by:

$$C_{eff} = \frac{3\pi(1-\nu_m)}{Z(1-\phi)G_m} \left[\frac{A_0}{\pi R^2} \text{erf} \left(\frac{\pi R^2}{A_0} \frac{4P_e}{ZE'(1-\phi)} \sqrt{\frac{2}{\langle |\nabla h|^2 \rangle}} \right) \right]^{-1/2} \quad (8)$$

Interestingly, at partial and full contact conditions (at high pressures), the bulk compressibility of a granular material composed of rough grains depends also on $A_0/\pi R^2$, which is not the case at low pressures (see Eq. (5)). If we set $A_0 = \pi b^2$ where b is the equivalent apparent contact radius, then $A_0/\pi R^2 = (b/R)^2$ holds. That is, the bulk compressibility is related to the ratio of apparent contact radius to the average grain radius (i.e., b/R) rather

than the absolute size of the grain. Thus, a special attention should be paid to the determination of b/R . Assuming that the porosity reduction with respect to a random packing of spheres with porosity of about 0.36 is caused by cementation deposited evenly on the grain surface, the estimate of b/R is given by $b/R = [2(\phi_0 - \phi)/3(1 - \phi_0)]^{0.5}$ where $\phi_0 \approx 0.36$ is the critical porosity for a dense random pack of spherical grains [16]. However, a part of the cement may not contribute to the initial contact radius, in particular when deposited in the equant pore space surrounded by neighbouring grains rather than on grain surface. This suggests that b/R estimated above may represent an upper bound.

3. Bulk compressibility of a rough granular material: an improved micromechanical approach

Three-dimensional microstructural observations on porous sandstones reveal that the pore space present in granular materials may be divided into three distinct categories: equant nodal pores at four-grain vertices, tubular pores at three-grain edges and narrow sheet-like throats (thin inter-granular cracks) at two-grain interfaces [70-72]. The bulk of the pore volume is contained in four- and three grain junctions compared to that of the narrow throats [73]. Cracks are mechanically more compliant than nodal and tubular pores [53], which are relatively stiff and insensitive to pressure. Typically, nodal and tubular pores are hydraulically connected by narrow cracks, exhibiting similar pore pressure in a fluid-saturated granular material. In the following, we develop an improved micromechanical model, which accounts for effects of different microscopic pore structures on the macroscopic bulk compressibility of a granular material.

3.1. Effect of narrow (compliant) cracks on bulk compressibility

A three-dimensional oblate spheroid with low aspect ratio < 0.1 is most commonly used to represent a crack-like void [55-60]. For a random dense packing of particles, the distribution of narrow cracks may be assumed to be homogenous and isotropic. In this case, the effective bulk compressibility when taking the interaction of cracks into account is nearly equal to that who are calculated under the assumption of non-interactions between cracks [57,61]. The effect of randomly distributed cracks on the effective bulk compressibility C_{eff} of an isotropic and homogeneous cracked material is expressed as [56,57,60,61,74]:

$$C_{eff} = C_m + \frac{16(1 - \nu_m^2)C_m}{9(1 - 2\nu_m)}\Gamma \quad (9)$$

where C_m and ν_m are the bulk compressibility and Poisson's ratio of the elastic solid matrix, respectively; Γ is the non-dimensional crack density defined as $\Gamma = \frac{1}{V} \sum_{i=1}^{N_c} r_i^3$ (or $\Gamma = \frac{N_c \langle r^3 \rangle}{V}$, the angle brackets symbolize an average) where r_i is the radius of i th crack and N_c indicates the number of cracks in a representative elementary volume V (REV). Typically, the progressive closure of cracks with increasing pressure results in a decrease in crack density, which is not explicitly given in Eq. (9). Hereafter, the pressure-dependent bulk compressibility of cracked material is linked to the evolution of microscopic parameters starting with the general closure law for a single crack when subjected to pressure.

Inter-granular crack surfaces are typically rough and characterized by a distribution of asperities. This suggests that for crack closure, the crack radius r and crack aspect ratio e ($e=w/r$, where w is the half length of short axis) may vary as a function of applied P_e . In particular, for very oblate spheroidal crack with smooth surface, the crack aspect ratio decreases linearly with increasing P_e whereas the crack radius remains unchanged, as formulated by $r(P_e) = r_0$ and $e(P_e) = e_0 - 2(1-\nu_m)P_e/\pi G_m$ [54,58,75] in which r_0 and e_0 are the initial crack radius and aspect ratio of a crack at the non-stressed state, respectively.

Considering that the only average crack radius defined in crack density Γ governs the bulk compressibility, all the inter-granular cracks are thought to have the initial “average crack radius” r_0 from a mathematical point of view. At non-stressed state, these cracks obey a statistical distribution of aspect ratios that depends on the initial microstructure of the porous granular materials. The number of cracks in a REV with aspect ratio lower than a given value e^* ($0 \leq e^* \leq e_{\max}$, $e_{\max}$ is the initial maximum value for aspect ratio of cracks) in a non-stressed state denoted by $N_c^0(e \leq e^*, P_e = 0)$ is given by:

$$N_c^0(e \leq e^*, P_e = 0) = N_0 \int_0^{e^*} f_e^0(e) de \quad (10)$$

where N_0 is the initial total crack number in a REV at the non-stressed state and $f_e^0(e)de$ corresponds to the fraction of cracks with an aspect ratio between e and $e+de$ at zero pressure (Fig. 2a).

Provided that the all inter-granular cracks evolve in a similar manner, they are expected to have a similar reduction in aspect ratio at a given P_e . In this case, the updated aspect ratio distribution function $f_e^{P_e}$ (the superscript represents the value of applied pressure) at a given P_e (Fig. 2a) can be deduced from the initial distribution f_e^0 at zero pressure by a horizontal shift β :

$$f_e^{P_e}(e) = f_e^0(e + \beta) \quad (11)$$

where β is the corresponding value of reduction of aspect ratio of a crack associated with P_e (i.e., $e(P_e) = e_0 - \beta(P_e)$). Since the cracks with initial aspect ratio lower than β (i.e., $e_0 < \beta$) are expected to be completely closed at a given P_e , they no longer contribute to the crack density and to the resulting bulk compressibility. Accordingly, in a REV the updated total number of cracks $N_c^{P_e}$ (the superscript represents the value of applied pressure) that still remain open at a given P_e can be derived as:

$$N_c^{P_e}(P_e) = N_0 \int_0^{e_{\max}-\beta} f_e^{P_e}(e) de = N_0 \int_0^{e_{\max}-\beta} f_e^0(e + \beta) de = N_0 \int_{\beta}^{e_{\max}} f_e^0(e) de \quad (12)$$

Furthermore, the crack density Γ can be expressed as a function of P_e by:

$$\Gamma(P_e) = \frac{1}{V} \sum_{i=1}^{N_c^{P_e}} r_i^3 = \frac{1}{V} N_0 r^3 \int_{\beta}^{e_{\max}} f_e^0(e) de \quad (13)$$

In addition, the total crack porosity ϕ_c at a given P_e can be approximately estimated by:

$$\phi_c(P_e) = \frac{1}{V} \frac{4\pi}{3} \sum_{i=1}^{N_c^e} e_i r_i^3 = \frac{1}{V} \frac{4\pi}{3} N_0 r^3 \int_{\beta}^{e_{\max}} f_e^0(e)(e - \beta) de \quad (14)$$

Differentiating Eq. (14) with respect to P_e and further simplifying it by use of Eqs. (13) and (14) yield:

$$\frac{d\phi_c}{dP_e} = \frac{3}{r} \frac{dr}{dP_e} \phi_c - \frac{4\pi}{3} \frac{d\beta}{dP_e} \Gamma \quad (15)$$

From Eq. (15) the resulting crack density Γ is then substituted into Eq. (9), the bulk compressibility C_{eff} of cracked material is then:

$$C_{eff} = C_m + \frac{4(1-\nu_m^2)C_m}{3\pi(1-2\nu_m)} \frac{dP_e}{d\beta} \left(\frac{3}{r} \frac{dr}{dP_e} \phi_c - \frac{d\phi_c}{dP_e} \right) \quad (16a)$$

For a given inter-granular crack, $e(P_e) = e_0 - \beta(P_e)$ results in $dP_e/d\beta = -dP_e/de$. Considering the equalities of $(dr/dP_e)/r = d \ln(r/r_0)/dP_e$ and $dP_e/d\beta = -dP_e/de$, Eq. (16a) can be rewritten as:

$$C_{eff} = C_m - \frac{4(1-\nu_m^2)C_m}{3\pi(1-2\nu_m)} \frac{dP_e}{de} \left[\frac{3d \ln(r/r_0)}{dP_e} \phi_c - \frac{d\phi_c}{dP_e} \right] \quad (16b)$$

In the following, we discuss this result in more detail for two different cases: a population of smooth-walled cracks and of rough-walled cracks, respectively.

3.1.1. Smooth cracks

For a crack with smooth surfaces, the corresponding closure law under pressure is given by: $r(P_e) = r_0$, $e(P_e) = e_0 - \beta(P_e)$ where $\beta(P_e) = 2(1-\nu_m)P_e/\pi G_m$ [54,58,75]. Substituting such a relation into Eq. (16a) and using $G_m = 3(1-2\nu_m)/[2(1+\nu_m)C_m]$, bulk compressibility C_{eff} of solid materials containing a random distribution of smooth cracks can be reduced to:

$$C_{eff} = C_m - \frac{d\phi_c}{dP_e} \quad (17a)$$

Eq. (17a) is consistent with the theoretical solution obtained by Walsh [53]. It suggests that in the presence of smooth cracks, the effective bulk compressibility only depends on decrease of crack porosity with increasing pressure, irrespective of total crack porosity.

3.1.2. Rough cracks

Experimental observations show that asperities are irregularly distributed over grain surfaces in sandstone [70]. In general, the closure of an inter-granular crack is controlled by the mutual indentation of asperities, which modifies the evolution of crack radius and of aspect ratio with pressure (i.e., crack closure law). The influence of ‘islands’ at partial contacts between surfaces on macroscopic stiffness may be mechanically incorporated into the framework of the crack density parameter by properly reducing the effective crack radius [60]. Indeed, the partial contacts between grains reduce the effective crack radius, leading to a reduction of crack compliance, as reflected in Eq. (16b). For a population of rough cracks embedded in a solid matrix, crack porosity, crack aspect

ratio and relative change of crack radius associated with pressure govern the effective bulk compressibility (see Eq. (16b)).

Following the approach initially developed by Johnson et al. [32], the open regions along rough surfaces may be regarded as penny-shaped internal cracks [25,29,76-78]. Specifically, the open region between two grains in contact with nominal contact area A_0 under non-stressed state may be regarded as a single crack with initial effective crack radius $r_0 = \sqrt{A_0/\pi}$ (Fig. 2b). As the external pressure is raised, asperities come in contact leading to an increase in real contact area A (A may be derived using contact mechanical models as described in Section 2) which is equivalent to a progressive decrease in crack radius (Fig. 2b). As a result, the updated effective crack radius r as a function of pressure is estimated as $\pi r^2 = A_0 - A$ or $r/r_0 = (1 - A/A_0)^{0.5}$. Given that $A/A_0 = \text{erf}(\lambda P_e)$ where λ is a constant (see Eq. (7) and assuming that the porosity ϕ and the coordination number Z change little with pressure), r/r_0 may be expressed by $r/r_0 = [1 - \text{erf}(\lambda P_e)]^{0.5}$. The crack closure pressure P_{cl} is defined as a characteristic hydrostatic pressure above which almost all cracks are closed [53], which may be estimated from the measurement of ultrasonic P- and S-wave velocities [54]. From the evolution of r/r_0 with increasing P_e (Fig. 3), $P_{cl} \approx 3/\lambda$ is suggested. In this case, the term of $d \ln(r/r_0)/dP_e$ in Eq. (16b) can be determined accordingly.

In contrast, precisely constraining the term of dP_e/de in Eq. (16b) is a big challenge because it may depend on the distribution of asperities on grain surfaces. However, at the initial low pressures (e.g., $r/r_0 > 0.9$ when $P_e < 0.17/\lambda$, see Fig. 3), only a few amount of asperities coming into contact between grains is expected to lead to the inter-granular cracks behaving under pressure like smooth cracks, suggesting that $\beta(P_e) = 2(1 - \nu_m)P_e/\pi G_m$ (closure law of smooth crack) may be still valid at low pressures. Hence, at low pressures Eq. (16b) may be approximated as:

$$C_{eff} \approx C_m - \frac{d\phi_c}{dP_e} - \frac{3\lambda \exp[-(\lambda P_e)^2]}{\sqrt{\pi} [1 - \text{erf}(\lambda P_e)]} \phi_c \quad (P_e < 0.17/\lambda) \quad (17b)$$

where $\lambda \approx 3/P_{cl}$ is suggested.

3.2. Effect of equant (stiff) pores on bulk compressibility

The most frequently used analogue of stiff pores is a three-dimensional spherical cavity or a two-dimensional tubular conduit with circular section [54,60,79,80]. But, these models generally overestimate the stiffness of equant pore space present in granular materials. Equant pores between adjacent spherical grains in granular materials typically have cusp-like corners, which geometrically differ from the blunt corner shape of spheroidal or circular cavities. Sharp or blunt corners at pores strongly influence the effective elastic properties of porous media [60,81].

Based on three-dimensional microstructural observations on granular materials [70-72], we approximate stiff pores as a set of conduits with cross sections of the space between three and four neighbouring spherical grains (Fig. 1, see the corresponding blue-shaded areas labelled by 2 and 1 in the inset of the micromechanical model,

respectively). The compressibility of tubular cavities with cross section of three cusp-like corners between three spherical grains in contact is given as [82]:

$$C_{pc}^{3T} = \frac{6(1-\nu_m)}{G_m} \quad (18a)$$

C_{pc} characterizes the compressibility of pore space measured in drained isotropic compression tests in which the pore pressure is remained constant while confining pressure is varied. Similarly, the compressibility of four cusp-like corners of tubular cavities resembling the nodal pores at four spherical grains contacts is written as [82]:

$$C_{pc}^{4T} = \frac{4(1-\nu_m)}{G_m} \quad (18b)$$

Using Betti's reciprocal theorem together with the theory of poroelasticity [83], the relationship between C_{pc} and effective bulk compressibility of porous material C_{eff} is given as:

$$C_{pc} = \frac{C_{eff} - C_m}{\phi_s} \quad (19)$$

where ϕ_s indicates the stiff porosity. Substituting Eq. (18a) and Eq. (18b) into Eq. (19), C_{eff} of the porous material containing a family of non-interacting tubular pores with cross section of three and four cusp-like corners, respectively, may be expressed by:

$$C_{eff}^{3T} = C_m + \frac{4(1-\nu_m^2)C_m}{1-2\nu_m}\phi_s \quad (20a)$$

$$C_{eff}^{4T} = C_m + \frac{8(1-\nu_m^2)C_m}{3(1-2\nu_m)}\phi_s \quad (20b)$$

Assuming that particles are distributed randomly within the granular material, the ratio of pores surrounded by three grains to pores surrounded by four grains is approximately 4:3. Accordingly, the weighted average of Eq. (20a) and Eq. (20b) (4/7 and 3/7, respectively) are adopted for simplicity to roughly assess the influence of stiff pore space on bulk compressibility, as expressed by:

$$C_{eff} = C_m + \frac{24(1-\nu_m^2)C_m}{7(1-2\nu_m)}\phi_s \quad (21)$$

Eq. (21) shows that the bulk compressibility is related to stiff porosity, insensitive to details of the microstructure.

3.3. Pressure-dependent bulk compressibility of a granular material

The superposition principle of elastic potential [56,61] allows expressing the pressure-dependent C_{eff} of granular materials by combining Eq. (16b) and Eq. (21) as:

$$C_{eff} = C_m - \frac{4(1-\nu_m^2)C_m}{3\pi(1-2\nu_m)} \frac{dP_e}{de} \left[\frac{3d \ln(r/r_0)}{dP_e} \phi_c - \frac{d\phi_c}{dP_e} \right] + \frac{24(1-\nu_m^2)C_m}{7(1-2\nu_m)} \phi_s \quad (22)$$

Considering Eq. (17b) at low pressures, Eq. (22) may be approximated as:

$$C_{eff} \approx C_m - \frac{d\phi_c}{dP_e} - \frac{3\lambda \exp[-(\lambda P_e)^2]}{\sqrt{\pi} [1 - \operatorname{erf}(\lambda P_e)]} \phi_c + \frac{24(1-\nu_m^2)C_m}{7(1-2\nu_m)} \phi_s \quad (P_e < 0.17/\lambda) \quad (23a)$$

where $\lambda \approx 3/P_{cl}$ is suggested. Combining the predicted C_{eff} at low pressures ($P_e < 0.17/\lambda$) using Eq. (23a) and at high pressures ($P_e \geq P_{cl}$) using Eq. (21) allow us to interpolate the bulk compressibility at intermediate pressures based on the suggested power law function fitting [1,4,9] or exponential function fitting [54].

In particular, if all cracks have smooth surfaces, Eq. (22) reduces to:

$$C_{eff} = C_m - \frac{d\phi_c}{dP_e} + \frac{24(1-\nu_m^2)C_m}{7(1-2\nu_m)} \phi_s \quad (23b)$$

4. Experimental program

In an effort to evaluate the different theoretical estimates of bulk compressibility of a porous granular material, we performed hydrostatic compression tests on Bentheim sandstone samples under drained and unjacketed conditions.

4.1. Sample materials and experimental setup

For testing, we selected Bentheim sandstone, which is a porous, homogenous and isotropic reservoir sandstone composed of ~96.5 % quartz, ~2 % feldspar and ~1.5 % clay [84]. The diameter of well-sorted and mostly rounded to sub-rounded quartz grains varies from 0.050 to 0.550 mm with a mean diameter of about 0.20mm. We determined connected porosity of our samples at ambient conditions of about 0.233 by measuring the net weight of dry and water-saturated samples. The microstructure of grain packing and pore space in Bentheim sandstone shows large pores and microcracks visible at grain-grain contacts (Fig. 4). Scanning electron microscopy (SEM) images reveal that grain boundaries are rough (amplitude < 1 μm) and partially open with a spacing of several hundred nanometers (Fig. 4).

Cylindrical specimens were drilled from the same block with a standard size of 50 mm diameter and 100 mm length. Samples were dried in an oven at constant 50 °C for at least one day. The samples were placed in a rubber jacket to isolate from the confining (oil) medium.

To monitor the deformation of samples under hydrostatic pressure, two pairs of orthogonally oriented electrical resistance strain gauges were glued onto the sample surface on opposite sides in axial and circumferential directions (see Fig. A1a in Appendix A). The volumetric bulk strain ε_b was determined from the expression $\varepsilon_1 + 2\varepsilon_3$, in which the axial strain ε_1 was calculated from the average values of two vertically oriented strain gauges and $2\varepsilon_3$ represents the sum of tangential strain values measured by two strain gauges oriented in a circumferential direction.

In addition, twelve P-wave and four polarized S-wave piezoelectric sensors were attached directly to the surface of the samples in order to simultaneously measure the ultrasonic velocities (1 MHz) along different paths across the samples (see Fig. A1b in Appendix A). This allows monitoring the change of sample elastic properties as a function of applied pressure.

4.2. Drained andunjacketed compression tests

A servo-hydraulic 4600 kN loading frame (MTS) equipped with a pore pressure system (Quizix 6000) (see Fig. A1c in Appendix A) was used to perform two hydrostatic compression tests at drained conditions with constant pore pressure for the bulk compressibility of Bentheim sandstone. Dry samples were first evacuated with vacuum pump at about 1 MPa confining pressure for 12 hours, and afterwards, samples were saturated with distilled water at constant pore pressure of 2 MPa and confining pressure of 2.8 MPa for several hours. Subsequently, the confining pressure was raised to 190 MPa at a loading rate of 1 MPa/min while the pore pressure was kept constant at 2 MPa. The drained fluid volume signifying the changes in volume of interconnected pore space during loading was continuously monitored with an accuracy of about $\pm 0.001 \text{ cm}^3$. The permeability of Bentheim sandstone is about $9 \times 10^{-13} \text{ m}^2$ at ambient pressure, which ensures completely drained conditions during hydrostatic compression tests.

To determine the bulk modulus of rock matrix, we conducted an ‘unjacketed’ test by using a jacketed sample and simultaneously increasing confining pressure and pore pressure at similar rates. To avoid any leakage of pore fluid, a small pressure difference between confining pressure and pore pressure was maintained constant at 3 MPa during loading up to a maximum confining pressure of 35 MPa. The bulk compressibility of the solid matrix can be calculated from the slope of a volumetric strain-confining pressure curve [83].

4.3. Experimental Results

4.3.1. Volumetric strain and ultrasonic wave velocities

In drained hydrostatic compression tests the volumetric strain of the bulk sample (ε_b) increases nonlinearly at confining pressures below about 60 MPa, followed by linear growth at higher pressures (Fig. 5a). The irreversible (plastic) strain of about 0.1 % was observed after unloading, only accounting for ~ 6 % of overall bulk strain. This suggests that the Bentheim sandstone is mainly deformed elastically in the range of applied pressure. The volumetric strain of the pore space (ε_p) is estimated from the ratio of the drained fluid volume ΔV_p to the volume of initial pore space V_p^i at the non-stressed state (i.e., $\varepsilon_p = \Delta V_p / V_p^i$). Calculated ε_p data as well as measured P- and S-wave velocities shows a rapid non-linear increase with effective pressure up to about 60 MPa, and an almost linear increase at higher pressures (Fig. 5b). Small scatter of wave velocities propagating along different directions suggests that the mechanical behaviour of Bentheim sandstone is largely isotropic when subjected to hydrostatic pressure.

4.3.2. Total porosity and crack porosity

The total porosity ϕ at any loading stage is estimated using $\phi = \phi_i(1 - \varepsilon_p)/(1 - \varepsilon_b)$, where ϕ_i is the initial porosity. The measured total porosity shows a small non-linear drop from initially ~ 0.233 to about ~ 0.226 at $P_e = 60 \text{ MPa}$, and a linear decrease at higher pressures (Fig. 6a). We assume that total porosity may be represented approximately by a combination of compliant cracks and stiff pores. The initial crack porosity (ϕ_c) constitutes

only a very small fraction of the initial total porosity and decreased rapidly to zero from about 0.47 % at $P_e = 1$ MPa (Fig. 6b). Beyond the closure of cracks, stiff porosity (ϕ_s) deforms linearly with increasing hydrostatic pressure (Fig. 6a). Below the crack closure pressure, crack porosity is obtained by subtracting the linear extrapolation of stiff porosity from total porosity.

4.3.3. Bulk compressibility and Poisson's ratio of solid matrix

The result of the 'unjacketed' compression test shows that the bulk compressibility of the solid matrix of Bentheim sandstone does not vary with applied pressure, yielding a constant value of $C_m \approx 0.031 \text{ GPa}^{-1}$. This corresponds to a bulk modulus of $K_m \approx 32.4 \text{ GPa}$ for the solid matrix (Fig. 7a), which is about 11 % lower than the bulk modulus of quartz (36.5 GPa) [14].

We assume a Poisson's ratio of quartz minerals ($\nu_m \approx 0.085$) [14] for the solid matrix of Bentheim sandstone because it is almost pure-quartz assemblage. From this and the measured bulk compressibility of rock matrix, we estimate shear modulus (G_m) and Young's modulus (E_m) of the rock matrix to be about 37.2 GPa and 80.7 GPa, respectively.

4.3.4. Bulk compressibility of bulk sample

The drained bulk compressibility C_{eff} of the sample may be determined from the local slope of ε_b plotted versus effective pressure ($C_{eff} = d\varepsilon_b/dP_e$) (Fig. 5a). This is expected to equal the bulk compressibility of dry Bentheim sandstone [14]. To minimize digitization noise, we first smoothed raw data by applying a moving average window of 21 points with a sampling rate of 2 Hz and afterwards fitted a high order polynomial to the entire pressure-volumetric strain curve. The resulting estimates of C_{eff} reduce drastically with increasing effective pressure from $\sim 0.30 \text{ GPa}^{-1}$ at $P_e = 1 \text{ MPa}$ to $\sim 0.085 \text{ GPa}^{-1}$ at $P_e \approx 60 \text{ MPa}$, and almost remains constant at higher pressures (Fig. 7b).

5. Discussion

5.1. Application of the extended contact model to describe bulk compressibility of a granular material

5.1.1. Bulk compressibility of a granular material predicted by contact models

With increasing hydrostatic pressure, porosity decreases and more grains are brought into contact [9-11]. We estimated the dependence of coordination number Z on porosity using $Z(\phi) = 24 \exp(-2.57\phi) - 0.371$ [11]. In our tests, Z is found to be almost independent of applied pressure, changing from ~ 12.8 to ~ 13.2 over the entire pressure range applied.

We compared the bulk compressibility of Bentheim sandstone predicted by continuum contact models (see Appendix B and Appendix C for details on Hertz model and Digby's model, respectively) with a prediction of the contact model presented here and our experimental data (Fig. 8a). The experimentally measured values of ϕ , G_m , C_m and ν_m are used in the model calculation. In smooth contact models, the only unknown parameter that we can adjust to fit experimental data is initial contact radius ratio b/R , where b is the initial contact radius between grains. Apparently, the bulk compressibility predicted by smooth contact models does not agree well the experimental data (Fig. 8a), which may be due to ignoring asperities distributed on grain surfaces.

In our developed contact models, two parameters ($\sqrt{\langle |\nabla h|^2 \rangle}$ and $A_0/\pi R^2$) in Eq. (8) need to be constrained in order to match the experimental bulk compressibility properly across the entire range of applied pressure. Using Eq. (8) based on least square regression, the best fit for bulk compressibility ($\sqrt{\langle |\nabla h|^2 \rangle} = 3.5 \times 10^{-3}$ and $A_0/\pi R^2 = 0.28^2$) with increasing pressure is in good agreement with experimental data at $P_e > 30$ MPa, but slightly deviates at low pressures. Here, we found that the relation between measured bulk compressibility of Bentheim sandstone and external pressure approximately follows a power law at low pressure less than 20 MPa with a pressure exponent lying between -1/3 and -1/2 (i.e., $C_{eff} \propto P_e^{-1/2.6}$ in Fig. 8b). In the following, we discuss the two basic microstructural parameters ($A_0/\pi R^2$ and $\sqrt{\langle |\nabla h|^2 \rangle}$) for Bentheim sandstone in detail.

5.1.2. Evolution of real contact area between rough grains

Evolution of real contact area A with pressure between rough grains affects bulk moduli of porous granular aggregates [8,13,15,17-19]. The evolution of contact area between rough grains may be estimated from experimentally measured bulk moduli using Eq. (1). Clearly, the experimentally-derived estimates of the normalized real contact area $A/\pi R^2$ varying with changing pressure may be divided into three stages (Fig. 9). At relatively low $P_e < 10$ MPa, the real contact area increases almost linearly with increasing external pressure up to about 0.03. This may result from purely elastic contact response of rough surfaces [42,43,69,85-87]. At intermediate contact pressures (i.e., in second stage), change of $A/\pi R^2$ with pressure deviates from linearity indicating gradual and slow growth in real contact area (Fig. 9). This could be attributed to the coalescence and interaction between contact zones associated with different asperities [40-42,68,69]. High local stresses developing at contacting asperities may cause yielding of the material [43,48,86]. At higher pressures, plastic yielding of asperities may result in complete contact for $P_e > 60$ MPa. Beyond complete contact pressure, $A/\pi R^2$ remains at a constant value of about 0.078 and is unaffected by increasing pressure (i.e., in third stage). The resulting constant bulk compressibility at high hydrostatic pressures is also observed on other granular materials [54, 88]. The numerical simulation on consolidation (sintering) process of powder particles shows that the growth of necks between particles at the early stage had a significant effect on the elastic modulus due to a rapid increase in real contact area [3]. However, once the neck size reached a critical value, the elastic modulus remained unchanged [3]. This evolving process for elastic modulus is similar to our observation.

In addition, we estimate $A/\pi R^2$ using smooth contact models at different initial contact radius ratios ($b/R = 0, 0.05$ and 0.28 , respectively) (Fig. 9). Smooth contact models underestimate the evolution of $A/\pi R^2$ with pressure for low initial contact ratios (e.g., $b/R = 0$ and 0.05). In contrast, almost constant value of real contact area predicted for $b/R = 0.28$ is in good agreement with experimental data at the elevated pressure > 60 MPa. This suggests that the contact of rough grains at pressures above a critical value at which full contact across the grain interface is established may be captured by continuum contact mechanics. This also implies that apparent contact area A_0 between rough grains is expected to remain almost unaffected by pressure once b/R is initially large (e.g., $b/R > 0.2$ for Bentheim sandstone). Using experimentally-derived $A/\pi R^2$ for Bentheim sandstone, the initial b/R between rough grains may be estimated to be about 0.28. Using $b/R = [2(\phi_0 - \phi)/3(1 - \phi_0)]^{0.5}$ [16], the upper bound of the initial ratio b/R was computed to be ~ 0.36 . Our estimated $b/R \approx 0.28$ for Bentheim sandstone meets this criterion.

5.1.3. Grain surface roughness

The roughness of grain surfaces and interfaces affects the evolution of real contact area A with pressure (see Eq. (4) and Eq. (7)). Note that $\sqrt{\langle |\nabla h|^2 \rangle}$ is a statistical measure of roughness (rms of surface gradients referred to the mean height of a grain surface). We provide the evolution of normalized real contact area $A/\pi R^2$ with pressure using Eq. (7) at the different roughness values (Fig. 9).

For a roughness of 6×10^{-3} (i.e., $\sqrt{\langle |\nabla h|^2 \rangle} = 6 \times 10^{-3}$), the predicted $A/\pi R^2$ agrees well with the experimental data at $P_e > 50$ MPa, but shows a small difference at lower pressures. Since Eq. (6) is found to be precise near full contact [45-50], the roughness of grains for Bentheim sandstone is estimated to be approximately 6×10^{-3} . In addition, we also predict the variation of $A/\pi R^2$ with pressure using Eq. (4) at two different values of $\kappa/\sqrt{\langle |\nabla h|^2 \rangle} = 500$ and 250 (Fig. 9). Considering that an asperity-based model (i.e., $\kappa = \sqrt{2\pi}$) is more appropriate to describe initial contact behaviour at very low pressures [42,68,69], we infer that the expected roughness of grains using asperity-based model at low pressure is about $(2\pi)^{0.5}/500 \approx 5 \times 10^{-3}$, which is close to the estimate using Eq. (7).

SEM images of polished surfaces at grain-grain contacts for Bentheim sandstone indicate that surface topography is very difficult to characterize analytically (Fig. 4). The polished surface profile allows evaluating the rms surface height change along a single direction (i.e., partial derivative of the mean height profile). After determination of the mean height plane of asperities along the grain interface, the estimated $\sqrt{\langle |\nabla h|^2 \rangle}$ is about of the order of 10^{-2} , and roughly consistent with the predicted values above.

5.2. Application of the improved micromechanical model to describe bulk compressibility of a granular material

For Bentheim sandstone deformed at hydrostatic pressures, the pressure-dependent stiff porosity (ϕ_s) and crack porosity (ϕ_c) are experimentally determined (see Section 4.3.2 above). Assuming smooth cracks within Bentheim sandstone, we estimate pressure-dependent bulk compressibility based on Eq. (23b). Although the predicted value at $P_e < 20$ MPa overestimates the compressibility, it agrees very well with the experimental data above the crack closure pressure of about 60 MPa (Fig. 10a). This suggests that our micromechanical model populated by tubular pores with cross-section of three and four cusp-like corners analogous to equant (stiff) pore spaces provides reasonable estimates of the bulk compressibility of granular materials at elevated pressures.

When $P_{cl} = 60$ MPa was taken, the bulk compressibility at low pressures $P_e \leq 3$ MPa was predicted using Eq. (23a). Although the predicted bulk compressibility at $P_e = 1$ MPa is significantly smaller than the corresponding measured value, this gap is narrowed dramatically with increasing P_e (Fig. 10b). In particular, the predicted and measured values of bulk compressibility are compatible at $P_e = 3$ MPa ($C_{eff} = 0.28$ GPa⁻¹ and 0.20 GPa⁻¹, respectively). At $3 < P_e < 60$ MPa, the interpolating results using power law function is more promising than using exponential function, consistent with the widely observed a power-law decay of the bulk compressibility versus pressure [1,4,9]. Compared to the smooth crack model, the consideration of rough cracks is more accurate to predict the bulk compressibility of Bentheim sandstone at intermediate pressures.

5.3. Comparison of contact mechanics and micromechanical models

Both contact and micromechanical models for describing pressure dependence of granular materials have been found to agree with experimental results, suggesting that there may exist an inherent correlation between the two approaches.

As initially developed by Johnson et al. [32], non-contacting regions between rough surfaces can be considered as parallel penny-shaped internal cracks along the interface plane [25,29,76-78]. For a granular aggregate consisting of rough grains, contact models consider deformation of contact areas (asperities) between two rough grains with increasing pressure in an effort to link contact microstructure to macroscopic mechanical properties. In contrast, micromechanical models focus on the mechanical closure of non-contact regions between two rough grains (i.e., inter-granular cracks) and shrinkage of large pores surrounded by several neighbouring grains (Fig. 1). The resulting decrease in the overall porosity including cracks and pores is expected to strengthen the bulk stiffness. Both contact and micromechanical models for Bentheim sandstone predict crack closure pressure of about 60 MPa. From the contact model, we find that prior to complete contact pressure, the real contact area increases rapidly with pressure, giving rise to significant decrease in bulk compressibility. Beyond full contact, the further evolution of contact area might be modelled by the continuous contact mechanics. Based on the micromechanical model, the progressive closure of compliant inter-granular cracks as a result of an increase of real contact area between rough grains causes the macroscopic bulk compressibility to reduce substantially. Above crack closure pressure, the remaining equant pore space surrounded by neighbouring grains is largely insensitive to pressure, with almost no further change in bulk compressibility. The predicted bulk compressibility for Bentheim sandstone indicates that the contact model seems to be more convenient to give relatively accurate results.

6. Conclusion

In this paper, we investigated the pressure-dependent bulk compressibilities of granular materials based on the contact mechanics and micromechanics, respectively.

The contact microgeometry between two rough grains is generally divided into real contact area and non-contact area, and the former essentially governs bulk compressibility of a granular material. Our extended contact model for a granular material composed of rough grains indicates that the bulk compressibility depends on hydrostatic pressure following a power law relationship with an exponent of $-1/2$ at low pressures, but deviates at intermediate pressures. Beyond full contact pressure, the bulk compressibility remains almost unchanged. Alternatively, in an attempt to incorporate general microstructural characteristics of pore space in granular materials, a micromechanical model has been proposed that accounts for different pore geometries within granular materials affecting macroscopic elastic properties. Narrow (compliant) inter-granular cracks are approximated by three-dimensional oblate spheroidal cracks with rough surfaces whereas the equant (stiff) pore space surrounded by three and four neighbouring grains is modelled as a combination of tubular pores with cross-section of three and four cusp-like corners. At low pressures, progressive closure of rough-walled cracks with increasing pressure considerably reduces effective bulk compressibility of a granular aggregate. Above the crack closure pressure, bulk compressibility stays almost constant with increasing pressure.

Both contact mechanics and micromechanical models link macroscopic mechanical properties of granular materials to microstructural characteristics, and are in good agreement with experimental results on Bentheim sandstone. Our improved models may provide a better understanding of pressure-dependent macroscopic mechanical properties of porous granular materials associated with microstructural deformation processes, such as in the field of compaction or consolidation of powder particles under pressure.

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Appendix A. Experimental setup

Fig. A1. (a) Sensor map presenting positions of P-wave piezoelectric transducers (P1 to P12), S-wave sensors (S_{H1} , S_{H2} , S_{V1} and S_{V2}) and strain gages. Half of the sensors were used for ultrasonic transmission (indicated by orange labels). (b) Photograph of a sample assembly with sensors and Neoprene jacket. (c) Schematic diagram of the experimental apparatus.

Appendix B: Hertz model (initial contact radius $b = 0$)

The effective elastic properties of spherical particle packings mainly depend on normal and tangential contact stiffnesses of a two-particle combination. As for a random unconsolidated sphere packing, the effective bulk compressibility can be theoretically expressed as [8]:

$$C_{eff} = \frac{12\pi R'}{Z(1-\phi)S_n} \quad (B.1)$$

where

$$\frac{1}{R'} = \frac{1}{R_1} + \frac{1}{R_2} \quad (B.2)$$

R_i and ϕ are the radii of two spheres (if they have different elastic properties) and the porosity, respectively; Z denotes coordination number (the average number of contacts per sphere), S_n is the normal stiffness defined as the ratio of a normal force increment to shortening distance between centres of two spheres.

In the case of the normal compression of two identical spheres with same radius R , the normal stiffness S_n , the mutual approach between centres of two spheres δ and contact radius a can be written as:

$$S_n = \frac{\partial F}{\partial \delta} = \frac{2aG_m}{1-\nu_m} \quad (B.3)$$

$$\frac{\delta}{2} = \frac{a^2}{R} \quad (B.4)$$

$$a = \left[\frac{3FR}{8G_m} (1-\nu_m) \right]^{1/3} \quad (B.5)$$

where G_m and ν_m are the shear modulus and the Poisson ratio of the grain material, F is normal force acting between two particles.

When the hydrostatic pressure P_e is applied to a random and identical sphere packing, the relation between P_e and F is constrained by:

$$F = \frac{4\pi R^2 P_e}{Z(1-\phi)} \quad (B.6)$$

Substituting Eqs. (B.6, B.5, B.3 and B.2) into Eq. (B.1), the effective bulk compressibility of random packing of identical spheres is then given as:

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$$C_{eff} = \left[\frac{Z^2(1-\phi)^2 G_m^2}{18\pi^2(1-\nu_m)^2} P_e \right]^{-1/3} \quad (B.7)$$

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It is clearly found that the pressure-dependent bulk compressibility of random packing of unconsolidated spherical grains follows $P_e^{-1/3}$ law. Due to the fact that a dense random packing of identical spheres has a porosity of about 36 %, the Hertz model is appropriate to describe the pressure dependence of elastic properties of granular material with relatively high porosity in the range of 36 ~ 40 %.

Appendix C. Digby's model (initial contact radius $b > 0$)

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Digby [15] analytically solved the problem of interaction of a random packing of identical and smooth spherical particles that are initially bonded together across small, flat and circular regions. If the neighbouring grains with same radius R are initially bonded across region of contact radius b , the applied normal force F should satisfy the relation as:

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$$F = \frac{4G_m}{3(1-\nu_m)} \frac{\sqrt{a^2 - b^2}}{R} (2a^2 + b^2) \quad (C.1)$$

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where G_m and ν_m are the shear modulus and Poisson ratio of grain, respectively; a is the varied contact radius dependent on external pressure. Clearly, in the case of $b=0$, it reduces to Hertz relation (see Eq. (B.5)). Substituting Eq. (B.6) into Eq. (C.1), one can obtain the following equation for the normalized contact radius a/R :

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$$\frac{a}{R} = \sqrt{\left(\frac{x}{R}\right)^2 + \left(\frac{b}{R}\right)^2} \quad (C.2)$$

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where x/R satisfies the cubic equation below

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$$\left(\frac{x}{R}\right)^3 + \frac{3}{2}\left(\frac{b}{R}\right)^2\left(\frac{x}{R}\right) - \frac{3\pi(1-\nu_m)P_e}{2Z(1-\phi)G_m} = 0 \quad (C.3)$$

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In addition, the correlation between the mutual approach between centres of two spheres δ and contact radius a is constrained by:

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$$\frac{\delta}{2} = \frac{a(a^2 - b^2)^{1/2}}{R} \quad (C.4)$$

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Combining Eqs. (C.2) and (C.3) with Eqs. (B.1) and (B.3), one can implicitly determine the effective bulk modulus of granular media as a function of hydrostatic pressure P_e and the initial normalized cemented radius b/R .

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Fig. 1. Schematic illustration of describing the mechanical response of general granular materials to hydrostatic pressure using contact mechanics and micromechanics, respectively.

Fig. 2. (a) Illustrative diagram of variation of aspect ratio distribution function with increasing pressure. The aspect ratio distribution function $f_e^{P_c}(e)$ at P_c may be deduced from the initial aspect ratio distribution $f_e^0(e)$ at zero pressure by horizontal transition β . Note that β is varied as a function of P_c . (b) Conceptual cross-section diagram of two rough grains in contact and the illustrative diagram of modelling the contact behaviour as a pressurised rough crack with varied effective crack radius r governed by the real contact area A between two rough grains. The real contact area A is composed of several individual asperities in contact (A' , A'' and A'''), lower than apparent contact area A_0 . The relative change of effective crack radius r/r_0 with increasing pressure is roughly estimated by $r/r_0 = \sqrt{1 - A/A_0}$. Note that the smooth surface of the crack in the third inset is only used for schematic representation and thus does not represent the real geometry of a rough crack.

Fig. 3. (a) The relative change of real contact area (A/A_0) with increasing effective pressure P_e follows an error function. (b) The derived relative change of effective real contact radius (r/r_0) with increasing effective pressure P_e . The crack closure pressure P_{cl} is approximated as $P_{cl} \approx 3/\lambda$. Note that $P_e = 0.17/\lambda$ results in $r/r_0 = 0.9$.

Fig. 4. (a-b) Thin-section optical micrographs of a natural granular material (Bentheim sandstone composed of almost pure quartz) impregnated with blue epoxy. Narrow inter-granular cracks at grain-grain contact and large pore space surrounded by several neighbouring grains are well interconnected forming a continuous network. (c-d) Scanning electron microscope (SEM) images of polished surfaces of rough grains in contact in Bentheim sandstone. Grain boundaries are rough (amplitude lower than 1 μm) and partially open with a spacing of several hundred nanometers.

Fig. 5. (a) Measured volumetric strain of bulk sample (ϵ_b) with increasing P_e in the first drained test. The unloading part is shown by dashed curve. (b) Calculated volumetric strain of pore space (ϵ_p) as well as measured P- and S- wave velocities as a function of P_e in first drained test. The error bar for P-wave velocities indicates standard deviation of P-wave velocities along axial and radial directions whereas the error bar for S-wave velocities denotes the standard deviation of horizontally and vertically polarized S- waves. The second drained test displays the similar results and is not shown here for simplicity.

Fig. 6. (a) Evolution of total porosity (ϕ) with increasing P_e in drained compression tests. (b) Determined crack porosity (ϕ_c) as a function of P_e by subtracting linear evolution of stiff porosity (ϕ_s) with pressure from total porosity (ϕ). The error bar indicates the maximum and minimum measurements of the two drained compression tests.

Fig. 7. (a) Evolution of volumetric strain of bulk sample (ε_b) with pressure variation in an unjacketed test. (b) Experimentally determined bulk compressibility (C_{eff}) of Bentheim sandstone with increasing P_e . The error bar indicates the maximum and minimum measurements of the two drained compression tests.

Fig. 8. (a) Comparisons of experimentally determined bulk compressibility (C_{eff}) of Bentheim sandstone as a function of P_e with corresponding predictions using smooth contact models and our improved contact models, respectively. The parameters of $b/R=0.26$ in Digby's model and $\sqrt{\langle |\nabla h|^2 \rangle} = 3.5 \times 10^{-3}$ and $A_0/\pi R^2 = 0.28^2$ in Eq. (8) are determined from the best fitting for C_{eff} over the entire range of pressure applied based on least square regression. (b) Experimentally determined C_{eff} with increasing P_e plotted in a double-logarithmic coordinate. A power law function ($C_{\text{eff}} = A \times P_e^B$) is used to fit experimental C_{eff} at $P_e < 20$ MPa with the parameter of $B = -1/2.6$.

Fig. 9. Experimentally derived evolution of real contact area normalized by the cross-section area of grain ($A/\pi R^2$) between two rough grains as a function of P_e using Eq. (1) and corresponding predictions by smooth contact models and rough contact models, respectively.

Fig. 10. (a) Comparisons of experimentally determined bulk compressibility with the prediction from micromechanical model using Eq. (23b). Note that Eq. (23b) assumes that all cracks have smooth surfaces. (b) The predicted bulk compressibility (red dots) using Eq. (23a) at low pressures ($P_e \leq 3$ MPa) when crack closure pressure $P_{\text{cl}} = 60$ MPa is taken. Using the predicted values of bulk compressibility at $P_e \leq 3$ MPa and at $P_e \geq 60$ MPa, the values of bulk compressibility at intermediate pressures ($3 \text{ MPa} < P_e < 60 \text{ MPa}$) are interpolated based on the curve fitting of the exponential function ($C_{\text{eff}} [\text{GPa}^{-1}] = 1.7 \times \exp(-0.7 \times P_e [\text{MPa}]) + 0.076$, $R^2=0.99$) and power law function ($C_{\text{eff}} [\text{GPa}^{-1}] = 0.86 \times (P_e [\text{MPa}])^{-1.11} + 0.064$, $R^2=0.99$), respectively.