

On comminution and yield in brittle granular mixtures

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Abstract

The mechanics of granular mixtures are pivotal in many industrial applications. Unravelling the relation between yielding and comminution, the action of mechanically induced grain size reduction, in confined mixture systems is a common and open challenge. This paper attacks this problem by adopting the breakage mechanics theory, which was originally proposed for single mineral materials. We present an extension to the theory that allows predicting: (1) the yielding pressure in granular mixtures, (2) the yield pressure increase/hardening with increasing breakage, and (3) the evolution of the grain size distributions of the separate species—all of these novel capabilities are tested and validated with experiments. Of particular appeal is the finding that the average yielding pressure is a simple generalized mean with an exponent $-3/2$ of the yielding pressures of the homogeneous components.

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1. Introduction

Granular systems composed of particles with dissimilar properties are found in a wide variety of fields. In mining operations, for example, granules composed of different mineral species are liberated from the parent geomaterial during fragmentation (Lynch, 1977). Similar processes occur in aggregate manufacturing, where large slabs of rock are broken into dissimilar grains with custom crushers (Kranz, 1979; Guimaraes et al., 2007). In natural resources, the extraction of water and hydrocarbons from heterogeneous formations is often accompanied by a rise in intergranular stress that can lead to the release of migratory fragments generated by the damage induced on softer grains (Koprulu, 2007). In turn, the migratory fragments can clog the formation. The food industry seeks to optimize the packaging, storage, and bulk delivery of dissimilar granular foods, such as grains, seeds, and cereals. Optimization demands predicting the quality degradation that different grains may suffer due to the package confinement; this prediction should be based on knowledge of the components' resistance against confinement as independent homogeneous systems. In environmental engineering, high-pressure granular filters are engineered by artificially mixing low-strength porous grains with stronger grains that provide stability to the assemblage and thus reduce the potential for porous grain damage.

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Encountering natural sand sediments composed of grains with different mineralogy (e.g., quartz, feldspar, and mica mixtures in desert regions; calcium carbonate and quartz mixtures in tropical coasts) is common in geology and geotechnical engineering practice (Way, 1973; Terzaghi et al., 1996). The structural loading of mixed sediments can induce more damage to weak grains than to stronger grains (Leleu and Valdes, 2007); in turn, dissimilar damage leads to behaviour alterations. The current state of the art in geotechnical engineering, however, involves determining engineering properties experimentally, ignoring any knowledge of the mineralogy and mineral composition heterogeneity of the granular sediment. Clearly, optimal engineering demands seeking solutions that do not rely on repetitive experimental testing. Instead, the goal should be to predict mixture properties given the properties of the independent components. Pursuing this goal is, in fact, the heart of this paper. To achieve this, the theory of breakage mechanics (Einav, 2007a), is applied extended and validated against experimental results.

2. Breakage mechanics for homogeneous systems

Breakage mechanics is only briefly reviewed here: the end-product is presented in the form of a set of master equations. The focus in this paper is the derivation of an extended theory that is meant to capture the behaviour of granular mixtures at high stresses. The derivation of the master equations of the homogeneous theory of breakage mechanics can give necessary mathematical and physical information (e.g., statistical micromechanics, energy balance, and particle scale mechanisms) and can be found elsewhere (Einav, 2007a, b).

Let x be an extensive statistical variate, a function of the grain size D . The average of this variable may be expressed using the grain size distribution (gsd) $p(D)$:

$$X \equiv \langle x \rangle = \int p(D)x(D) dD \quad (1)$$

where the gsd is a density function, therefore satisfying $\int p(D) dD \equiv 1$.

The breakage mechanics equations for *homogeneous* brittle granular systems may be collected:

$$p(D) = p_0(D)(1 - B) + p_u(D)B \quad (2)$$

$$\mathfrak{B} = 1 - \langle D^2 \rangle_u / \langle D^2 \rangle_0 \quad (3)$$

$$\Psi \equiv (1 - \mathfrak{B})\psi_r(\varepsilon) \quad (4)$$

$$E_B = E_B(\varepsilon) = -\frac{\partial \Psi}{\partial B} = \mathfrak{B}\psi_r(\varepsilon) = \frac{\mathfrak{B}\Psi}{1 - \mathfrak{B}} \quad (5)$$

$$\sigma = \frac{\partial \Psi}{\partial \varepsilon} = (1 - \mathfrak{B})\psi_r'(\varepsilon) \quad (6)$$

$$E_B^* = E_B(1 - B) \quad (7)$$

$$\Phi_B = \delta E_B^* \geq 0 \quad (8)$$

$$y_B = E_B(1 - B)^2 - E_c \leq 0 \quad (9)$$

where B , the breakage, weighs using Eq. (2) the relative distance of the current gsd $p(D)$ from the initial and ultimate gsd $p_0(D)$ and $p_u(D)$; \mathfrak{B} in Eq. (3) is referred to as the criticality proximity parameter and is measured using the initial and ultimate second-order moments of the gsd, i.e., using $\langle D^2 \rangle_0$ and $\langle D^2 \rangle_u$, i.e., how far the initial distribution is from the ultimate distribution (which for most practical purposes can be postulated as a fractal distribution); the macroscopic specific Helmholtz free energy potential Ψ is connected to the breakage and macroscopic strain tensor ε based on Eq. (4); the stress-like conjugate to the breakage is referred to as the breakage energy E_B , and is expressed in various ways using Eq. (5) by differentiating Ψ ; the real Cauchy stress σ is, on the other hand, the conjugate of the strain, derived using Eq. (6). It is then convenient to define the residual breakage energy E_B^* using Eq. (7), to express how much energy is left in the system for breaking

particles, at any given moment, even after some breakage has already occurred. The loss in the residual breakage energy δE_B^* , resulting from the incremental breakage is postulated by Eq. (8) to equate to the increment of breakage dissipation Φ_B . Combining Eqs. (7) and (8) and integrating gives the breaking-yield condition y_B in Eq. (9), where E_c is introduced as a constant of integration, referred to as the critical breakage energy constant (Einav, 2007c).

The corresponding explicit form of the breakage dissipation is given as

$$\Phi_B = \frac{\sqrt{E_c E_B(\varepsilon)}}{1 - B} \delta B \geq 0 \quad (10)$$

where $E_B(\varepsilon)$ is explicit function given by the fourth term in Eq. (5). Using the above equations, Einav (2007c) proposed that homogeneous granular systems break in compression according to

$$p_{cr} = p_r \sqrt[3/2]{\frac{3E_c K^*}{4\vartheta p_r}} \quad (11a)$$

where p_{cr} was referred to as the critical yielding pressure, or critical comminution pressure; p_r is the reference pressure taken as 1 kPa, a small but sufficient pressure for the particles to be confined and for the system to be considered using continuum mechanics principles; K^* is the Hertzian dimensionless constant in the power-law relation of the bulk modulus $K = K^* p_r (p/p_r)^m$, with m being the Hertzian constant, taken as 1/2.

Acknowledging the limits of the Hertzian contact model in correctly predicting the stiffening effect from rising coordination numbers with pressure, to a first-order energy considerations suggest that the bulk modulus is inversely proportional to the porosity (e.g., Walton, 1987), i.e., that $K^* = K_e(1 + e)/e$, therefore:

$$p_{cr} = p_r \sqrt[3/2]{\frac{3E_c K_e}{4\vartheta p_r} \left(\frac{1 + e_{cr}}{e_{cr}} \right)} \quad (11b)$$

where e_{cr} is the void ratio of the granular pack once the critical yielding pressure is reached. In the following we are interested in deriving a similar formula but for the mixture problem. Note that the exponent $-3/2$ in the above relation equals ‘ $m-2$ ’, with the coefficient m taken as 1/2. This coefficient may differ in practice for some reasons (e.g., rising coordination number, angular particles, etc.), roughly between $m = 1/3$ and $2/3$.

3. Breakage mechanics for N-phases heterogeneous systems

The first goal of this paper is to generalize the breakage mechanics formulation for mixtures, generally containing N distinct phases, where the separate phases correspond to different types of particle species. As in other continuum mixture theories (Truesdell, 1961; Green and Naghdi, 1969; Hashin, 1983), the different species are assumed to exist everywhere in the representative material element. Let $p^{(i)}(D)$, $p_0^{(i)}(D)$, and $p_u^{(i)}(D)$, be the current, initial and ultimate gsd of phase i . Let $f^{(i)}$ be the solid-volume fraction of phase (i). The theory is meant to capture the behaviour of brittle granular mixtures: particles cannot undergo substantial deformations prior to crushing, and therefore do not loose solid volume. In other words, the solid-volume fractions of the various species remain constant during loading and unloading processes. Therefore, global mass conservation requires that for a system of rigid particles

$$\sum_{i=1}^N f^{(i)} = 1 \quad (12)$$

Alternatively, the equation above could have been specified directly in terms of mass fractions rather solid-volume fractions. The two options converge only when the specific gravity of the species is the same. However, the use of volume rather than mass is more relevant when describing brittle granular systems because it is the volume that directly carries the information of the grain size. Particle size (not mass) controls the spatial distribution and number of force chains that are to be transferred through the particular species and how much energy these particles store. Similarly, the definition of the gsd by mass or volume in using Eq. (2) for the

single phase materials would not affect the results because the specific gravity is essentially constant for the mineral and the two definitions converge.

Before we proceed, it is useful to define yet another type of average—different to the gsd average definition of Eq. (1)—that is based on the volume fractions. Therefore, if $Z^{(i)}$ is a variable representing phase (i), its mixture average is written as

$$\bar{Z} = \sum_{i=1}^N Z^{(i)} f^{(i)} \tag{13}$$

Let $B^{(i)}$ be the breakage in fraction i . In the mixture formulation we define Eq. (2) for the individual phases:

$$p^{(i)}(D) = p_0^{(i)}(D)(1 - B^{(i)}) + p_u^{(i)}(D)B^{(i)} \tag{14}$$

The average gsd of the entire mixture is given by weighing the individual distributions of the mixture using the phase fractions:

$$\bar{p}(D) = \sum_{i=1}^N f^{(i)} p^{(i)}(D) = \sum_{i=1}^N f^{(i)} (p_0^{(i)}(D)(1 - B^{(i)}) + p_u^{(i)}(D)B^{(i)}) \tag{15}$$

Free energy is an additive quantity, so that the overall specific mixture energy is simply the average of the specific energy of the separate phases, but taking the gradation into account:

$$\bar{\Psi} \equiv \sum_{i=1}^N \psi^{(i)}(\varepsilon, D) f^{(i)} p^{(i)}(D) \tag{16}$$

In Eq. (16), $\psi^{(i)}(\varepsilon, D)$ is the stored energy in an average particle with a size D in fraction i . Einav (2007a) proposed, and tested numerically, that on average, the energy that particles store scales linearly with their surface area. Considering spherical particles, we write

$$\psi^{(i)}(\varepsilon, D) = \frac{\psi_r^{(i)}(\varepsilon) D^2}{\langle D^2 \rangle} \tag{17}$$

Combining Eq. (17) with Eqs. (14) and (16), and defining the list of criticality proximity parameters for the various phases:

$$g^{(i)} = 1 - \langle D^2 \rangle_u^{(i)} / \langle D^2 \rangle_0^{(i)} \tag{18}$$

we write

$$\bar{\Psi} \equiv \sum_{i=1}^N \psi_r^{(i)}(\varepsilon) f^{(i)} (1 - g^{(i)} B^{(i)}) = \bar{\psi}_r(\varepsilon) - \sum_{i=1}^N \psi_r^{(i)}(\varepsilon) g^{(i)} f^{(i)} B^{(i)} \tag{19}$$

where we define

$$\bar{\psi}_r(\varepsilon) = \sum_{i=1}^N f^{(i)} \psi_r^{(i)}(\varepsilon) \tag{20}$$

Before any crushing occurs, $B^{(i)}$ is zero for any i , and the specific stored energy, in Eq. (19), is therefore given by Eq. (20). This suggests that given the solid volume fractions $f^{(i)}$ and an assumed rule for the instantaneous elasticity behaviour of the individual fractions via $\psi_r^{(i)}(\varepsilon)$, we should be able to get the macroscopic instantaneous elasticity of the entire mixture using Eq. (20).

Now let us move to the dissipation, subjecting attention to rate independent processes. We designate the mixture dissipation as $\bar{\Phi}_B$, and define the breakage energy in phase i using

$$E_B^{(i)} = \frac{\partial \bar{\Phi}_B}{\partial \delta B^{(i)}} \tag{21}$$

The mixture dissipation is the average of the dissipative power of the distinct phases (defined as a phase’s breakage energy times the incremental change of that phase’s breakage):

$$\bar{\Phi}_B = \sum_{i=1}^N f^{(i)} E_B^{(i)} \delta B^{(i)} \tag{22}$$

Combining Eqs. (19) and (22), and acknowledging energy conservation $\sigma \delta \varepsilon = \bar{\Phi}_B + \delta \bar{\psi}$, we write

$$\sigma = \frac{\partial \bar{\psi}}{\partial \varepsilon} = \bar{\psi}'_r(\varepsilon) - \sum_{i=1}^N f^{(i)} \mathfrak{g}^{(i)} \psi_r^{(i)}(\varepsilon) B^{(i)} \tag{23}$$

$$E_B^{(i)} = E_B^{(i)}(\varepsilon) = -\frac{\partial \bar{\psi}}{\partial B^{(i)}} = \mathfrak{g}^{(i)} \psi_r^{(i)}(\varepsilon) \tag{24}$$

Note that before any crushing, all $B^{(i)}$ are zero and $\sigma = \bar{\psi}'_r(\varepsilon)$. Also note that we may define the mixture breakage energy as

$$\bar{E}_B = \sum_{i=1}^N f^{(i)} E_B^{(i)} = \sum_{i=1}^N \mathfrak{g}^{(i)} f^{(i)} \psi_r^{(i)}(\varepsilon) \tag{25}$$

Finally, note that if the initial and ultimate gsd of all of the phases in the mixture are assumed similarly, the list of criticality proximity parameters satisfies $\mathfrak{g}^{(i)} = \mathfrak{g}$ for any i , so the above equation could be specialized by

$$\bar{E}_B = \mathfrak{g} \sum_{i=1}^N f^{(i)} \psi_r^{(i)}(\varepsilon) = \mathfrak{g} \bar{\psi}'_r(\varepsilon) \tag{26}$$

The key results, however, do not depend on following either Eq. (25) or (26), therefore for generality we continue with Eq. (25), i.e., for any $\mathfrak{g}^{(i)}$.

In writing Eq. (21), the relation $E_B^{(i)} = \partial \bar{\Phi}_B / \partial \delta B^{(i)}$ was deliberately used, rather than $E_B^{(i)} = \partial \Phi_B^{(i)} / \partial \delta B^{(i)}$. Our understanding is that the energy dissipation from a phase i , $\Phi_B^{(i)}$, should principally be coupled to the energy dissipation from any other phase $\Phi_B^{(j)}$ for $j \neq i$. Although the dissipation from the weaker phases may, in effect, start prior to the dissipation from the stronger phases, we seek an alternative, i.e., a rather more practical, yet physically motivated treatment that will allow us to define a single yielding point. The release of energy from the local collapse of force chains due to the initial crushing of weaker particles transmits the excess forces to the stronger particles; these forces may more rapidly than expected lead to the breakage of stronger particles. Coupling the dissipation contributions from the different phases, reflects this asymmetry, but more importantly, will allow us to proceed conveniently by deriving a single yield condition—which is more practical. The mathematical implications of this point will be clarified later, and the ability of our theory to predict experimental results will amplify the power of this approach.

The remaining ingredient in the theory requires postulating the precise dependence of the mixture dissipation on the phase dissipations. Let us start by postulating the following general structure:

$$\bar{\Phi}_B = \sqrt{\sum_{i=1}^N (\Phi_B^{(i)})^2 (f^{(i)})^n} \geq 0 \tag{27}$$

At this stage n is an unknown coefficient, but we will later prove that $n = -1$, and that the experimental data gathered later confirms its validity (no adjustments or iterations made). This postulate was taken before the experimental results. Applying Eq. (21) over Eq. (27) gives:

$$E_B^{(i)} = (f^{(i)})^n \frac{\Phi_B^{(i)}}{\Phi_B} \frac{\partial \Phi_B^{(i)}}{\partial \delta B^{(i)}} \tag{28}$$

In accord with Eq. (10), we assume that the phase-dissipation terms are given by

$$\Phi_B^{(i)} = \frac{\sqrt{E_c^{(i)} E_B^{(i)}(\varepsilon)}}{1 - B^{(i)}} \delta B^{(i)} \geq 0 \tag{29}$$

where $E_B^{(i)}(\varepsilon)$ is the fourth term in Eq. (24), an explicit expression of the strain. This is mathematically different from the term $E_B^{(i)}$, which holds true only when dissipation occurs.

Therefore, applying Eq. (28) gives

$$(f^{(i)})^{-n} E_B^{(i)} = \frac{\Phi_B^{(i)}}{\bar{\Phi}_B} \frac{\sqrt{E_c^{(i)} E_B^{(i)}(\varepsilon)}}{1 - B^{(i)}} \tag{30}$$

Rearranging and summing over the phases

$$\sum_{i=1}^N (f^{(i)})^{-n} \frac{(E_B^{(i)})^2}{E_B^{(i)}(\varepsilon) E_c^{(i)}} (1 - B^{(i)})^2 = \sum_{i=1}^N \frac{(\Phi_B^{(i)})^2 (f^{(i)})^n}{(\bar{\Phi}_B)^2} = 1 \tag{31}$$

Therefore, the mixture breaking-yield criterion is given by

$$\bar{y}_B = \sum_{i=1}^N (f^{(i)})^{-n} \frac{(E_B^{(i)})^2}{E_B^{(i)}(\varepsilon) E_c^{(i)}} (1 - B^{(i)})^2 - 1 \leq 0 \tag{32a}$$

and after omitting the strain ε , the reduced form is

$$\bar{y}_B = \sum_{i=1}^N (f^{(i)})^{-n} \frac{E_B^{(i)}}{E_c^{(i)}} (1 - B^{(i)})^2 - 1 \leq 0 \tag{32b}$$

The reason for distinguishing between the strain-dependent and independent forms of the yield functions in Eqs. (32a) and (32b) is that the flow rule evolution is defined based on the former (see Einav (2007a) for more detail).

Now we turn back to ask what the value of the coefficient n should be. The answer is simple. Consider a homogeneous granular pack in initial conditions, i.e., with $B^{(i)} = 0$ for any i , but look at this homogeneous pack as a heterogeneous mixture of N identical materials. Since $E_B^{(i)}$ and $E_c^{(i)}$ are now the same, they can come out of the summation in Eq. (32b). On the other hand, we may always chose any series of $f^{(i)}$ as long as the list satisfies Eq. (12). Therefore, in order that Eq. (32b) yields a similar result as the homogeneous solution in Eq. (9), n must be -1 . Only then $\sum_{i=1}^N (f^{(i)})^{-n} = 1$. In this case, the dissipation in Eq. (27) and the yield criterion in Eq. (32b) become:

$$\bar{\Phi}_B = \sqrt{(\Phi_B/f)^2} \geq 0 \tag{33}$$

$$\bar{y}_B = \frac{\bar{E}_B}{E_c} (1 - B)^2 - 1 \leq 0 \tag{34}$$

which justifies the use of the overhead bar on \bar{y}_B : the mixture yield condition is simply the arithmetic mean of the yield criteria of the separate species as homogeneous systems. On the other hand the mixture dissipation is a special square root mean of the species dissipation.

Note that although the yield criterion depends on the arithmetic mean of the $E_B^{(i)}$'s of the species, $E_B^{(i)}$ itself is non-linearly dependent on the pressure, and the i th specie's gsd and void ratio through Eqs. (23) and (24), as addressed in the next section.

4. Fracture criterion for N-phases heterogeneous mixtures

Einav (2007c) applied the breakage mechanics theory to derive the critical comminution pressure of isotropically compressed granular systems: Eq. (11) (i.e., the pressure at the commencement of grain crushing).

Analogous to Griffith's critical tensile strength for a crack embedded in a plate to be opened, Einav's formula describes the interrelation between three important aspects of the compressive fracture process in confined brittle granular matter: (1) the material, via the critical breakage energy E_c (an analogue to G_c in Griffith's expression) and bulk modulus K_e (an analogue to the Young's modulus E); (2) the pressure level p_{cr} (an analogue to Griffith's tensile strength σ_{cr}); and (3) the geometry of the particles given by the grains surface area through ϑ (an analogue to the initial crack length '2a' in Griffith's analysis). The critical comminution pressure was shown to depend on void ratio as well. The result of this addition is logical, suggesting that agglomerates break earlier if initially looser. This is in agreement with experimental results (e.g., Nakata et al., 2001). Our motivation in this article is to extend the analysis for granular mixtures. For that we follow the mixture formulation, and test the theoretical results against experimental data.

4.1. A theoretical formula

Before any crushing $B^{(i)} = 0$ in all phases. Therefore, the initial breaking-yield condition is given by

$$\bar{y}_B = \sum_{i=1}^N f^{(i)} \frac{\vartheta^{(i)} \psi_r^{(i)}}{E_c^{(i)}} - 1 \leq 0 \quad (35)$$

For a Hertzian model, we use

$$\psi_r^{(i)} = \frac{p_r}{(1-m)(2-m)K_e^{*(i)}} \left(\frac{p}{p_r} \right)^{2-m} \quad (36)$$

where $K_e^{*(i)} = K_e^{(i)}(1+e)/e$. Combining this with $m = 1/2$ and since the pressure corresponds to the mixture critical comminution pressure, we write

$$\psi_r^{(i)} = \frac{4p_r e_{cr}}{3K_e^{(i)}(1+e_{cr})} \left(\frac{p_{cr}}{p_r} \right)^{3/2} \quad (37)$$

Note that in the above the critical void ratio refers directly to the entire mixture in hand, rather to the species $e_{cr}^{(i)}$. The critical comminution pressure of phase i is defined by changing Eq. (11):

$$\left(\frac{p_{cr}^{(i)}}{p_r} \right)^{3/2} = \frac{3E_c^{(i)}K_e^{(i)}}{4\vartheta^{(i)}p_r} \left(\frac{1+e_{cr}}{e_{cr}} \right) \quad (38)$$

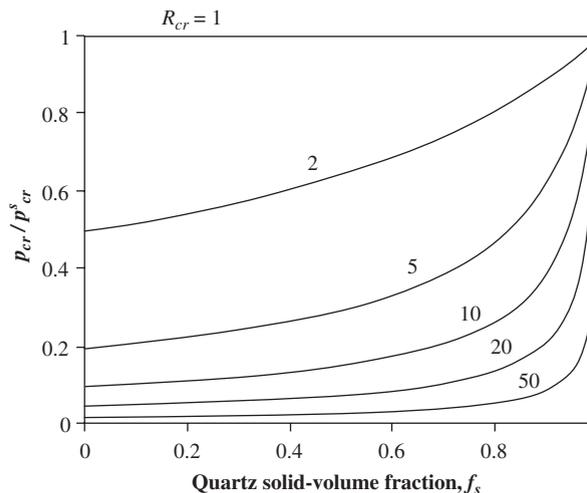


Fig. 1. R_{cr} -dependent variation in mixture critical pressure for mixtures of strong and weak particles. Note that mixtures with a high R_{cr} undergo a quicker reduction in strength with increasing weak grain content, i.e., with decreasing f_s .

Combining Eqs. (37), (38) with Eq. (35) enables us to derive the following formula for the mixture critical comminution pressure:

$$\bar{p}_{cr} = \sqrt[-3/2]{p_{cr}^{-3/2}} \quad (39)$$

Therefore, the average critical yielding pressure is simply the generalized mean with an exponent $-3/2$ (the Hertzian modulus minus two) of the separate critical phase yielding pressures of the homogeneous components. For example, for two-phase mixtures, the last equation becomes:

$$\bar{p}_{cr} = \sqrt[-3/2]{f_w(p_{cr}^w)^{-3/2} + f_s(p_{cr}^s)^{-3/2}} = p_{cr}^s((1 - f_s)R_{cr}^{3/2} + f_s)^{-2/3} \quad (40)$$

where f_w and f_s are the weak and strong phase volume fractions ($f_w + f_s = 1$), and $R_{cr} = p_{cr}^s/p_{cr}^w$ is defined as the critical comminution pressure ratio. Given an R_{cr} value, Eq. (40) provides a relation between the mixture critical comminution pressure and the strong phase volume fraction, as plotted in Fig. 1.

Note, the apparently simple Eqs. (39) and (40) are in fact rich with information. Through the application of the mixture average definition in Eq. (13), and the species' critical pressures in Eq. (38), the result of the overall yielding depends on the species' strength, stiffness, gsd , and porosity differences.

4.2. Formula validation using experimental results

The mixture formulation presented above was tested with experiments of dry sand crushing in constrained compression. Each specimen was constructed by mixing two sand species of dissimilar mineralogy but equal and uniform gradation. Similar grain shapes were used to hinder particle shape effects and to minimize the role of each species' relative density on the relative density of the prepared mixtures. In addition, we deliberately selected sands that could be separated with respect to mineralogy after testing; indeed, mineralogical separation yields complementary information about the granular breakage evolution of the different phases in the crushing assemblage. Set 1 consisted of mixed specimens composed of standard Ottawa sand with minimum and maximum void ratios $e_{min} = 0.52$ and $e_{max} = 0.72$ composed entirely of strong quartz grains, and manufactured calcareous sand with $e_{min} = 0.56$ and $e_{max} = 0.8$ composed entirely of weak-grained calcite grains; all with maximum particle diameter $D_{max} = 0.85$ mm minimum particle diameter $D_{min} = 0.6$ mm. Set 1 specimens with quartz sand fraction $f_s = 0, 0.1, 0.3, 0.7, 0.9, \text{ and } 1$ were prepared with an initial void ratio e_0 ranging between 0.54 and 0.56 (with e_0 values closer to 0.56 for lower f_s specimens), and compressed to selected target stresses. After loading, the fragments were separated with respect to grain size by standard sieving and with respect to mineralogy by dissolving the calcareous grains retained on each sieve with hydrochloric acid. Set 2 consisted of mixed specimens composed of fine Ottawa quartz sand with $e_{min} = 0.55$ and $e_{max} = 0.74$, and beach sand from Panama with $e_{min} = 0.57$ and $e_{max} = 0.77$ composed entirely of medium-strength magnetite grains; all with maximum particle diameter $D_{max} = 0.3$ mm and minimum particle diameter $D_{min} = 0.25$ mm. Set 2 specimens with quartz sand fraction $f_s = 0, 0.2, 0.5, 0.8, \text{ and } 1$ were prepared with $e_0 = 0.6$ and compressed to selected target stresses. After loading, the fragments were separated with respect to grain size by standard sieving and with respect to mineralogy by subjecting the portion retained on each sieve to the magnetic force of a permanent magnet, thus separating the magnetic magnetite grains from the quartz grains.

The $f_s = 1$ and 0 specimens provided values of p_{cr} and e_{cr} for each phase. Recall that the theoretical construct developed in this paper is based on the formulation developed by Einav (2007c), which describes compressive fracture for granular assemblages confined isotropically. The specimens tested herein were loaded in constrained compression, for which it is customary to report a characteristic 'yield' stress σ_y , i.e., the stress that marks the point of maximum curvature of void ratio vs. $\log(\sigma_v)$ response, where σ_v is the vertical stress. Therefore, values of p_{cr} for the specimens tested herein were calculated as $p_{cr} = \sigma_y(1 + 2(1 - \sin \varphi))/3$, where the friction angle was assumed to be $\varphi = 35^\circ$.

Results for Sets 1 and 2 are presented in Fig. 2, wherein each data point denotes a single experiment. The theoretical predictions are shown for comparison, highlighting the ability of the theory to forecast the sensitivity of the critical-pressure to variations in f_s ; in particular the quasi-linear and non-linear variations that correspond to bi-mixtures of moderately contrasted and highly contrasted strength. The theory's

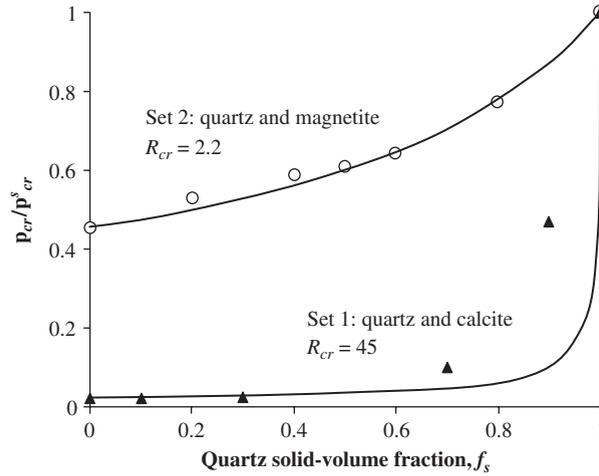


Fig. 2. Theoretical predictions of the critical yielding pressures in granular mixtures (Eq. (40)) compared against experimental data. Note that the theory does not present any fitting besides fixing the points that correspond to homogeneous specimens (i.e., $f_s = 0$ and 1).

deviation from the experimental trend increases as the difference in the grain species' strengths of the mixture increases (i.e., as the R_{cr} increases), and seems to occur particularly when f_s is beyond the percolation threshold, i.e., when f_s is about more than half. A possible explanation is linked to the fact that confined granular systems tend to transmit contact forces via distinct strong and weak force chain networks (Vardoulakis, 1989; Radjai et al., 1998). When f_s exceeds the percolation threshold, a strong force chain may travel in its entirety through the stronger phase, potentially without crossing any weak particle. This non-local effect would tend to cause destructive forces to deviate from the weak phase; thus leading to enhanced overall strength that a local theory cannot capture. The use of complimentary discrete element method (e.g., Cundall and Strack, 1979) studies could be used to extend the experimental data, by investigating mixtures with more pronounced strength, stiffness, gsd, and porosity contrasts. Nevertheless, the successfulness of the comparison is emphasized by underlining that the only points that were known *a priori* are those that pertain to homogeneous specimens (i.e., the $f_s = 0$ and 1 specimens).

5. The relation between breakage, grading and pressure in N-phases heterogeneous mixtures

In the previous section we focused on establishing a relation for the critical comminution pressure for granular mixtures. This section focuses on establishing relations for the evolution laws of the phases' breakage variable; in this way we are able to predict not only how the overall mixture gsd evolves, but also how the separate gradations evolve.

5.1. Theoretical analysis

The reason for distinguishing between the strain-dependent and independent forms of the yield functions in Eqs. (32a) and (32b) will now come into play. This is because the flow rule, defining the evolution of the fraction breakage variables, is given by differentiating the former. Now we can define the evolution of the various fraction breakage terms as the associated flow rule to this surface:

$$\delta B^{(i)} = \lambda \frac{\partial \bar{y}_B}{E_B^{(i)}} = \lambda f^{(i)} \frac{2E_B^{(i)}}{E_B^{(i)}(\epsilon)E_c^{(i)}} (1 - B^{(i)})^2 = \lambda f^{(i)} \frac{2}{E_c^{(i)}} (1 - B^{(i)})^2 \quad (41)$$

The factor '2' is the outcome of rigorously distinguishing, and differentiating Eq. (32b). The ratio between the i th and j th increment breakage terms is independent of the non-negative multiplier λ :

$$\frac{\delta B^{(i)}}{\delta B^{(j)}} = \frac{f^{(i)} E_c^{(j)}}{f^{(j)} E_c^{(i)}} \left(\frac{1 - B^{(i)}}{1 - B^{(j)}} \right)^2 \quad (42)$$

For example, in bi-mixtures we have

$$\frac{\delta B^w}{\delta B^s} = \frac{f^w}{f^s} \frac{E_c^s}{E_c^w} \left(\frac{1 - B^w}{1 - B^s} \right)^2 \tag{43}$$

This equation tells us that in predominantly strong specimens, the weak material would initially break at a slower rate than in predominantly weak specimens. In 50–50% specimens

$$\frac{\delta B^w}{\delta B^s} = \frac{E_c^s}{E_c^w} \left(\frac{1 - B^w}{1 - B^s} \right)^2 \tag{44}$$

Initially when $B^w = B^s = 0$:

$$\frac{\delta B^w}{\delta B^s} = \frac{E_c^s}{E_c^w} \tag{45}$$

This means that weak granules will start breaking quicker because E_c^s is bigger than E_c^w . As the weak granules keep breaking, ‘ $1 - B^w$ ’ becomes smaller than ‘ $1 - B^s$ ’ so the net effect of ‘ $E_c^s(1 - B^w)^2$ ’, in Eq. (44), would be to try and catch up with ‘ $E_c^w(1 - B^s)^2$ ’.

Eq. (43) is integrable, giving

$$\frac{B^w E_c^w}{(1 - B^w) f^w} = \frac{B^s E_c^s}{f^s (1 - B^s)} \tag{46}$$

Eq. (40) was written for the critical conditions of breakage initiation. It can be modified to account for the increasing pressure:

$$\bar{p}_{cr} = p_{cr}^s (f^w (1 - B^w)^2 R_{cr}^{3/2} + f^s (1 - B^s)^2)^{-2/3} \tag{47}$$

Solving Eq. (46) for the breakage in the strong and weak phases, and combining with Eq. (47), gives the pressure as a function of the breakage in the weak and strong phases:

$$p(B^w) = \left((p_{cr}^s)^{-3/2} \left(\frac{(1 - B^w) E_c^s f^w}{B^w E_c^w f^s + (1 - B^w) E_c^s f^w} \right)^2 f^s + (p_{cr}^w)^{-3/2} (1 - B^w)^2 f^w \right)^{-2/3} \tag{48}$$

$$p(B^s) = \left((p_{cr}^s)^{-3/2} (1 - B^s)^2 f^s + (p_{cr}^w)^{-3/2} \left(\frac{(1 - B^s) E_c^w f^s}{B^s E_c^s f^w + (1 - B^s) E_c^w f^s} \right)^2 f^w \right)^{-2/3} \tag{49}$$

Generalization gives the solution for N -phases material. The macroscopic pressure versus the n -phase-breakage is given by

$$p(B^{(i)}) = \left(\sum_{n=1}^N (p_{cr}^{(n)})^{-3/2} f^{(n)} \left(\frac{(1 - B^{(i)}) E_c^{(n)} f^{(i)}}{B^{(i)} E_c^{(i)} f^{(n)} + (1 - B^{(i)}) E_c^{(n)} f^{(i)}} \right)^2 \right)^{-2/3} \tag{50}$$

5.2. Theory validation using experimental results

Separation post-testing with respect to mineral composition yielded grain size distributions for the weak and strong fractions in each specimen tested. The experimental distributions were compared to the predictions prescribed by Eqs. (48) and (49). It should be noted, however, that distinctive experimental difficulties were encountered for both quartz-calcite specimens and quartz-magnetite specimens. For quartz-calcite specimens ($R_{cr} = 45$) with $f_s = 0, 0.1, \text{ and } 0.3$, i.e., predominantly weak grained, the imposed loading produced an agglomeration of calcite fragments; leading to the formation of a congruent block jammed inside the compression cell. The agglomeration of calcite fragments was not quantifiable. Furthermore, all attempts to remove the block from the cell were probably accompanied by grain damage. Therefore, the validity of the

grain size distributions obtained for these specimens was deemed questionable. In contrast, quartz–calcite specimens with $f_s = 0.7, 0.9,$ and 1 were easily removed from the cell as unbounded fragments. The differences observed are attributed to the preferred failure modes of the constituent minerals, as dictated by their crystallographic structure: quartz fractures conchoidally (and therefore abruptly) due to a lack of cleavage planes, whereas calcite fails along parallel planes of low molecular strength. It appears that upon loading, calcite fragments suffer gradual contact yielding, which causes an interlocking effect in specimens where the calcite content is more than 70% (i.e., $f_s \leq 0.3$). Indeed, granular packs composed of calcareous grains yield gradually as opposed to packs of quartz grains, which yield abruptly; similar observations are found for single grains loaded between platens (McDowell, 2002). For all quartz–magnetite specimens ($R_{cr} = 2.2$), difficulties arose in the separation process: a small fraction of the magnetite grains became demagnetized post-crushing; therefore, a slight magnetite contamination of the quartz fraction retained on each sieve occurred, thereby rendering an underestimation of magnetite breakage and overestimation of quartz breakage. In addition, the

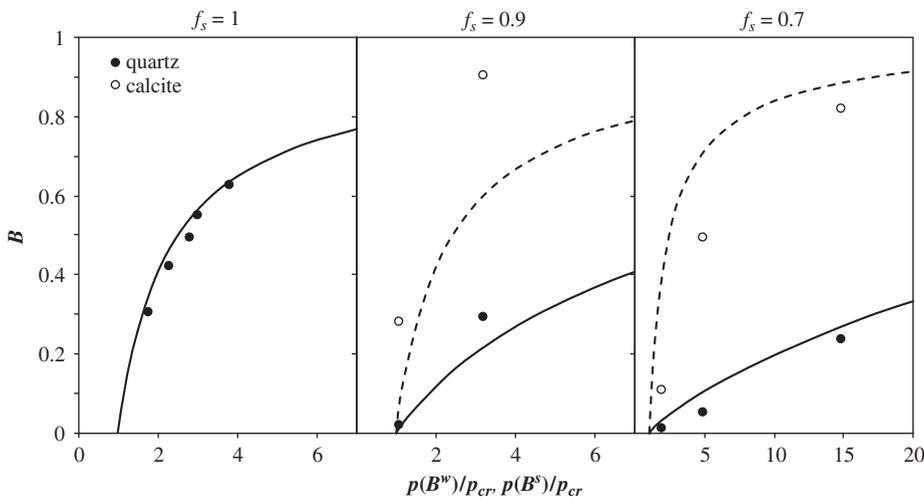


Fig. 3. Experimental and predicted breakage evolutions for predominantly strong-grained specimens with $R_{cr} = 45$ and quartz fraction f_s . Predictions with Eqs. (48) and (49) are shown with dashed and solid lines, respectively.

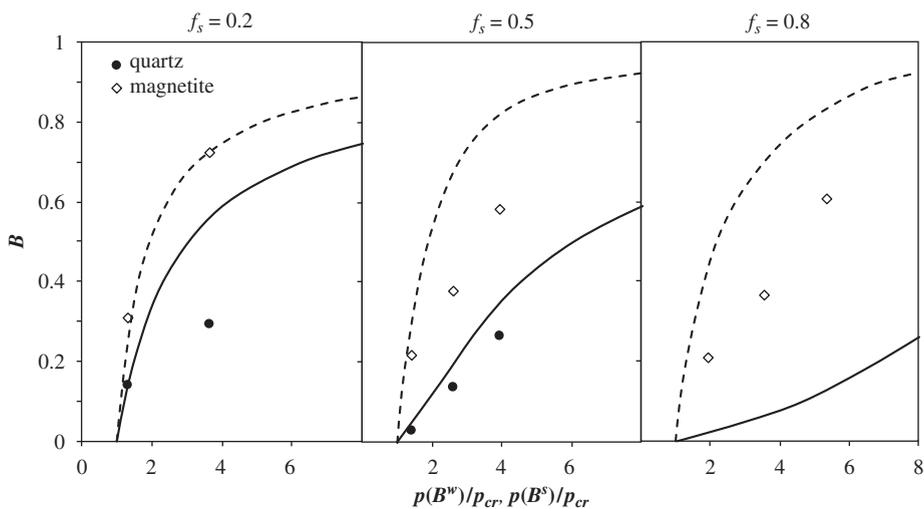


Fig. 4. Experimental and predicted breakage evolutions for predominantly strong-grained specimens with $R_{cr} = 2.2$ and quartz fraction f_s . Predictions with Eqs. (48) and (49) are shown with dashed and solid lines, respectively (breakage data for quartz for $f_s = 0.8$ not presented).

amount of the quartz material that was used for the $f_s = 0.8$ experiments was insufficient to yield reliable particle size distribution data from sieve analyses, given that (1) magnetite has a much larger specific gravity than quartz, and (2) small specimens were required to reach reasonably high \bar{p}_{cr}/p_{cr}^s values within the functional limits of the compression cell and the loading system.

The use of the theoretical Eqs. (48) and (49) requires the critical constants of the weak and strong phases E_c^w and E_c^s . These were calculated by employing the measured critical pressures p_{cr} of the homogeneous tests described in Section 4.2 and Eq. (11). For quartz–calcite specimens, the values of $K_e^{(w)}$ (calcite) and $K_e^{(s)}$ (quartz) were obtained using the instantaneous elastic stress–strain curve and were found to be 166 and 1350, respectively. Given $\vartheta^{(w)} = \vartheta^{(s)} = 0.85$, we found E_c^w (calcite) = 33.3 kPa and E_c^s (quartz) = 1621 kPa. For quartz–magnetite specimens, we found $K_e^{(w)}$ (magnetite) = 2750, E_c^w (magnetite) = 20.0 kPa, K_e^s (quartz) = 900, and E_c^s (quartz) = 1700 kPa. Fig. 3 shows the evolution of the species' breakage parameter $B^{(j)}$ with increasing pressures for quartz–calcite specimens with $f_s = 0.7, 0.9$, and 1 loaded to $\bar{p}_{cr}/p_{cr}^s > 1$. The theoretical predictions obtained with Eqs. (48) and (49) are also shown. The corresponding data and predictions for quartz–magnetite specimens ($R_{cr} = 2.2$) with $f_s = 0.2, 0.5$, and 0.8 are presented in Fig. 4 (note that the breakage values for the quartz fractions of the $f_s = 0.8$ specimens are omitted from Fig. 4 due to the difficulties discussed above). The quality of all of the predictions are emphasized by noting that there are no elements of fitting besides the departure points, which are, by themselves being predicted in Fig. 2.

6. Conclusions

Granular mixtures are ubiquitous in nature and man-made systems. The mixture strength is dependent on the properties of the constituent species. The theory of breakage mechanics, which was originally proposed for homogeneous granular materials (Einav, 2007a), was extended to the analysis of heterogeneous granular mixtures, taking into account the strength of the independent homogeneous species. We found that the mixture critical comminution pressure is a simple generalized mean with an exponent $-3/2$ (the Hertzian power coefficient of $m = 1/2$ minus 2) of the critical comminution pressures of the homogeneous components. We also furthered the mixture theory to permit predicting the breakage evolution of the combined and separate species in a granular mixture. The capability of both components of the theory, i.e., the prediction of critical comminution pressure and the prediction of breakage evolution for dissimilar constituent species, was confirmed with experiments, highlighting the completeness of the mixture formulation in particular and the power of breakage mechanics in general.

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