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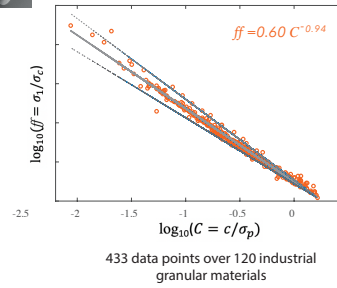
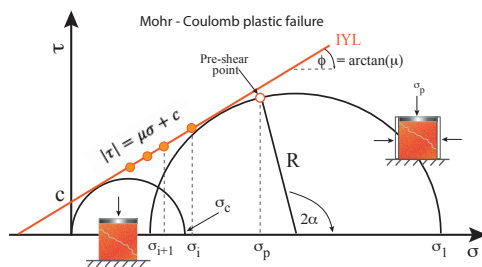
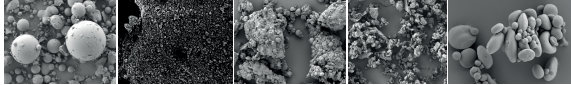
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Graphical Abstract

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Highlights

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- A data-driven model based on consolidation measurements of over 120 industrial powders employing Mohr-Coulomb theory.
- Principal Component Analysis shows that the bulk friction does not contribute to data variance.
- Granular rheology in the quasi-static regime is governed and defined by adhesive interactions.
- Fluctuations in the quasi-static regime are maintained by boundary conditions imposed at the boundaries of the quasi-static zones.

A phenomenological law for complex granular materials from Mohr-Coulomb theory

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Abstract

Manipulating powders still entails some clumsy and/or risky operations even now in the middle of the fourth industrial revolution. This is because there is a lack of well-understood theory about granular matter due to its ravelled complexity. However, granular matter is the second most handled material by man after water and is thus ubiquitous in daily life and industry only after water. Since the eighteenth century, mechanical and chemical engineers have been striving to manage the many difficulties of grain handling, most of which are related to flow problems. Many continuum models for dense granular flow have been proposed. Herein, we investigated Mohr-Coulomb failure analysis as it has been the cornerstone of stress distribution studies in industrial applications for decades. This research gathers over 130 granular materials from several industrial sectors, as varied as cement and flour, including raw materials, food, pharmaceuticals, and cosmetics. A phenomenological law derived from the yield locus and governed exclusively by one dimensionless number from adhesive interactions has been found. Surprisingly, and in contrast to the common perception, flow in the quasi-static regime is actually independent of the friction, the packing fraction and any other grains/bulk intrinsic properties. The simplicity and accuracy of the model are remarkable in light of the complex constitutive properties of granular matter.

Keywords: Granular materials, Powders, Mohr-Coulomb, Flowability, Principal component analysis

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1. Introduction

While classical mechanics describes the motion of material objects since Newton’s work in 1687, and Newtonian fluids via Navier-Stokes equations since 1845, the behaviour of granular materials in motion has yet to be successfully described. The Nobel prize winner Pierre Giles de Gennes stated that granular matter in 1998 was at the level of solid-state physics in 1930 [1] and ever since we have been searching for a unified vision to describe the granular flow. Most efforts seem to have focused on finding constitutive relations that could govern granular flow through dimensionless variables such as: a coefficient of friction $\mu = \tau/\sigma$ as the shear τ , to normal, σ , stress ratio for cohesionless grains; the packing fraction ϕ ; the inertial number $I \equiv \dot{\gamma}d/\sqrt{\sigma/\rho_s}$ where d and ρ_s are grains diameter and density, sheared at $\dot{\gamma}$ under a confining normal stress, σ ; an effective cohesion number $C^{eff} \equiv N_c^{eff}/(\sigma d^2)$ [2] with N_c^{eff} the ”effective adhesion”; and a granular temperature $\Theta \equiv \rho_s T/\sigma$ where T represents velocity fluctuations[3, 4]. The most popular constitutive relation is the $\mu(I)$ rheology [5] which loses accuracy when used for non-homogeneous shear flows. The most recent constitutive relations for granular rheology include adhesive interactions $\mu(I, C^{eff}) - \phi(I, C^{eff})$ [2] for which the authors also elucidate the importance of grains’ mechanical properties, or $\mu\Theta^p = f(I)$ [4] where p depends on the space dimension and $f(I)$ on particle information. All these remarkable works describe simple granular materials well but diverge when predicting “real-life” granular flows.

From a technological point of view, powder flow is essential to process feasibility-poor flow results in wastage, machine maintenance requirements, manufacturing downtime, and all the associated costs [6]. Several industrial and fundamental studies focus on the consolidation of granular media as it covers many technological fields [7, 8, 6, 9]. When one thinks of the vast cargoes of sand, cement, and wheat to be stored and transported, the importance of the relationship between consolidation and flowability becomes self-evident [10, 11, 12]. Whether we are dealing with a few grams or large installations, good flow behaviour means that the bulk solid will not consolidate during transport or storage, and it will flow out of a hopper, bins, or a silo due to gravity. Those who work with coarse or fine granular media such as powders know that powder flow is complex in nature [13, 14, 5]. The flow properties of granular materials have been found not to be an inherent property of the ma-

terial as this depends on the intrinsic and extrinsic properties of the grains, the equipment design, the processing conditions such as stress levels [15] and the environment [16, 17]. Even if the phrase “good or bad flow behaviour” as a qualitative statement can or seems to be easily described with the naked eye as when playing with dry sand at the beach or in the kitchen when pouring flour, the quantitative statement remains a physics and engineering challenge because of the lack of constitutive equations. Quantitative statements are only possible by using a physical characteristic of the bulk solid, which is attributed to be responsible for its flow behaviour [8, 16]. Thus, each flow device available on the market “estimates/quantifies” flowability from powders’ macroscopic properties using different dimensionless parameters [6, 15, 5]. The word “flowability” is usually misinterpreted as describing the material flow behaviour or is device-dependent as it represents the ability to flow in equipment under specific mechanical conditions [15, 16]. Although the compilation of these measurements is necessary in industrial development, it lacks a clear physical foundation [5, 6] and has been the subject of an overwhelming amount of literature.

Despite the advances in fundamental knowledge, the intriguing phenomena observed and described such as force chains, density waves or jamming [13, 14], none of the many physical postulates [18, 11, 19, 20, 21, 5, 4, 22] appear to have broad applicability to describe real-life granular materials which are also classified as complex, industrial, or cohesive. We seem to have reached a stage where the gap between fundamental science, mainly based on glass beads and sand, and the industry, based on many empirical but ingenious grain-based solutions, has once more become too great. Indeed, although the flow dynamic of monodisperse rigid cohesionless grains has been better assessed, the behaviour of more complicated granular materials such as geophysical matter, raw materials, food, pharmaceuticals, and cosmetics, to name but a few, is vastly less well understood.

Over the past years, the authors’ research group have dealt primarily with poor flow granular materials affecting product quality or process feasibility in many industrial applications. In this article, the inherent heterogeneous nature of granular matter is taken into account by analyzing the largest grain-based dataset we know to have been presented in the literature based on consolidation measurements. Many devices exist to quantify “flowability” from consolidation measurements. The first method employs compaction testers to link bulk density to flow behaviour through Hausner or Carr ratio [15, 16, 6]. A second employs shear testers to assess “flowability” from plastic

failure [10, 23, 11, 8]. This research focus on Mohr-Coulomb plasticity as the simplest and most widely used continuum description of granular matter in engineering. An original statistical analysis of the yield locus variables has been performed, leading to the definition of a dimensionless adhesion number to represent flow in the quasi-static regime irrespective of any other grain/bulk properties.

2. Theoretical background, flow in the quasi-static regime

When an elastic solid is subjected to normal stress, σ , the sample will deform as a function of the stress/strain answer of the material. For Hookean solids, the relationship between strain, γ , and stress, τ , is linear $\tau = G\gamma$ where G is the shear modulus. For viscous fluids, it is the rate of deformation, $\dot{\gamma}$, which is a function of the applied stress, with its simplest form for Newtonian fluids $\tau = \mu_f \dot{\gamma}$ where μ_f is the viscosity. A more complex situation emerges for granular matter as it consists of individual particles whose size, d , is larger than one micron, presenting a large variety of metastable states depending on the flow velocity [24]. If we study the grains as a continuum quasi-static medium, instead of forces between individual particles, the system can be described by forces, or stresses, on boundary areas of volume elements. At low shear, the material will deform elastically until it reaches a critical stress value, at which point the bulk yields into two rigid blocks that slide past each other separated by a narrow plastic zone. The Coulomb yield criterion describes this rigid-plastic failure [11]

$$\tau = \mu \sigma + c \tag{1}$$

where μ is a friction coefficient, and c represents bulk adhesive interactions. Both parameters depend on the material nature. The physical origins of the frictional properties of grains are still not clear.

As stresses are complicated mathematical objects, like a second-order tensor, that do not explicitly depend on the unit normal of any particular plane, a century after Coulomb's theory, Mohr popularized a graphical construction to represent the state of stress at a point [11]. To calculate flow from plastic failure, Jenike proposed a coaxial flow rule based on incompressibility and symmetry with respect to the principal stress planes—a flow rule is a constitutive law selected to reflect the overall behaviour of the material. The Mohr-Coulomb stress balance equations with the Jenike coaxial flow

rule of continuum plasticity (MCP) give rise to a dimensionless parameter known as the flow factor, ff , the ratio of the major principal stress, σ_1 , to the unconfined yield strength, σ_c (see Fig. 1) we can write

$$ff = \frac{\sigma_1}{\sigma_c} \text{ with } \sigma_1 = p + R \text{ and } \sigma_c = 2c \frac{\cos \phi}{1 - \sin \phi} \quad (2)$$

The larger the ff , the better the granular media flows [10]. This mathematical formulation is usually called the Mohr-Coulomb plasticity (MCP) and is the simplest, most sophisticated and most widely used model for granular materials in engineering.

For ideal Coulomb materials, all stresses are equal in magnitude, and thus σ_p is part of the IYL, while for low-cohesive samples, the IYL confounds with the EYL (see Fig. 1).

It should be noted that the Coulomb yield criterion is based on ideal Coulomb materials, which is an oversimplified picture of the behaviour of granular materials. Indeed, the Coulomb yield criterion “is a mathematical model and not a universal truth” [11]. The limits of the Coulomb yield criterion and their applicability are related to the fundamental assumption that the bulk can be treated as an ideal Coulomb material, meaning: *rigid-plastic continuous media which yields according to a Coulomb yield criterion*. Thus, highly cohesive samples (fine particles strongly agglomerate and difficult to fluidize [25]) are not part of the datasets studied in this work because they do not yield according to the Coulomb criterion.

3. Material and method

3.1. Granular materials

The samples are highly varied and include raw materials, inorganic materials, active principles, excipients, and several kinds of formulations from the Food, Pharmaceutical, and Cosmetic industries, including glidant-based formulations and 3D printing powders. Most of them submitted to confidential agreements (see DOREL repository for data properties). In dataset 1, about 40% of the samples were rigorously conditioned at 30% or 60% relative humidity.

In dataset 3 to bring to the fore the anisotropic nature of granular materials, six materials commonly used in industry were chosen: two grades of microcrystalline cellulose (MCC), wheat flour (WF), joint filler (JF), sericite

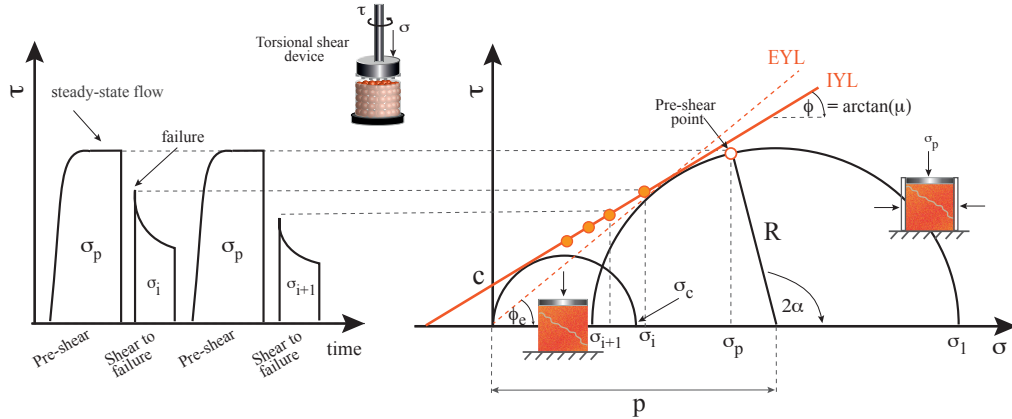


Figure 1: Schematic representation of the Shear Test with the graphical representation of the shear stress chart (left), the Shear Points (orange dots), Mohr Circle constructions (right), and the derived parameters. The Mohr circle constructions begin with plotting the pre-shear point and all the shear test points as normal(σ)/shear(τ) stress data pairs (orange dots). The internal yield locus, IYL, is the linear regression line (in orange) passing through the tests points until it meets the y-axis at $\sigma=0$, where c represents bulk adhesive interactions and ϕ the so-called angle of internal friction, μ . The construction of Mohr circles is centred on the x-axis. The minor circle is drawn by passing through the origin and tangent to the IYL. The non-zero intersection with the x-axis represents the maximum compressive stress that can act along a free surface, σ_c . The Major Mohr circle, centred at the point $(p,0)$ with radius R , is the circle tangent to the IYL that passes through the shear point. The greater intersection point with the x-axis defines the major principal stress, σ_1 . EYL is the effective yield locus, a straight line through the origin tangent to the Major Mohr circle, and ϕ_e is the so-called effective angle of internal friction.

(S) and glass beads (GB). WF is an artisanal T45 flour (Type 00, Euro 450 flour, or American Cake flour) obtained from the Poinignon flour mill (Fouigny, France) and was chosen to represent the food industry. MCC Avicel PH102 and PH105 (FMC Biopolymer) are common excipients used in the cosmetics and pharmaceutical industries. JF (MAPEI) is a gypsum-based material used in construction to grout tiles and finish walls. S is a mica powder-industry grade provided by a cosmetic enterprise. Finally, the GB, Spheriglass 3000-E (Potters Industries LLC), is used for highway safety marking. It should be noted, that the samples were conditioned under strict air humidity conditions, 30 and 60% RH as described in previous works [16],

and their water activity, A_w , was quantified.

3.2. Physical characterization techniques

Particle size distribution was measured in triplicate by laser diffraction in liquid media using a Mastersize 3000 (Malvern Instruments), ethanol was used as a dispersant to avoid dissolution. Particle morphology images were determined by SEM (Gemini 3, Zeiss) with a field-emission gun operating at 2 kV. Samples were placed on double-sided tape and then coated for 1 min with a gold-palladium layer using a Desk V - TSC (Denton Vacuum) under air purge. True powder density was determined in a Helium pycnometer (AccuPyc 1330, Micromeritics). All data presented correspond to the average of ten measurements obtained from one powder sample.

3.3. Flow behavior in the quasi-static regime

The experiments were carried out using a torsional shear tester: the FT4 powder rheometer from Freeman Technology with a 25mm x 10mL split vessel made of borosilicate glass. The slip is achieved by rotating the top of the bulk solid with a circular plate piston with a rough underside (23.5mm in diameter). This configuration may only be utilized with grains smaller than 1 mm to avoid friction with the piston's rough side (blade separation).

The shear-test procedure can be divided into four steps. The test starts with a mechanical conditioning step by establishing uniform stress into the powder bed using a blade. Once the sample is aerated, the granular media is subjected to normal stress, σ_p , using a vented piston allowing the trapped air within the powder to escape. When the consolidation cycle is over, the vessel splits leading to a bulk volume of 10 mL. Afterwards, the vented piston is replaced with the shear head. The third pre-shear step consists in reaching the steady-state shear stress by rotating at a fixed rate of 18°/min while maintaining the pre-consolidation stress. Then a zero torque level is established. The fourth, shear to failure step allows the IYL normal/shear stress data points to be determined (Fig. 1, orange dots). Plastic failure under consolidation is observed in the stress chart when the shear stress maximum value is reached and then subsequently reduced (stress chart Fig. 1, failure). The shear test points are obtained by shearing to failure at normal stress σ_i between 80% and 20% smaller than σ_p . Each of our routines contains five measurement points with a single pre-consolidation cycle at σ_p is carried out before each measurement point.

4. Results and Discussion

4.1. Physical characterization

Dataset 1 is composed of 287 data points from more than 100 different samples, from raw materials to several types of formulations. Samples mean size diameters varies from: 5 til 679 μm for the Sauter mean diameter ($d_{3,2}$) and from 10 to 736 μm for the Brouckere mean diameter ($d_{4,3}$). As for the ρ_p varies from 983 til 2810 kg/m^3 .

Dataset 2 is composed of 433 data points: the dataset 1 more 146 data points gathered from the scientific literature [26, 27, 28, 6, 29, 9, 17]. This will enlarge dataset 1 spectra by including heavy/fine samples (ρ_p 5800 kg/m^3 for $d_{3,2} \leq 15\mu\text{m}$ [6, 17]) and very cohesive samples such as calcium carbonate ($d_{3,2} = 4\mu\text{m}$ [9]).

Dataset 3 physical characterization is presented in detail in Table 1. SEM micrographs are shown in figure 2. All samples exhibit a polydisperse size distribution with densities from 1500 to 2900 kg/m^3 . In the Geldart classification for fluidization of granular materials, Avicel Ph 102 is A type (aeratable or homogeneously fluidizable), and Avicel PH 105, glass beads 3000E and Sericite are of type C (non-fluidizable). While due to their size, Joint filler and wheat flour are within the fuzzy boundary between both types.

Table 1: Physical properties of the samples in dataset 3.

Powder	Particle size distribution [μm]					True density [g/mL]	Geldart's classification
	d_{10}	d_{50}	d_{90}	d_{32}	d_{43}		
Avicel PH-102	35	111	225	122	69	1.55	A
Avicel PH-105	7	21	45	24	13	1.58	C
Glass beads	4	24	60	29	8	2.66	C
Wheat flour	17	83	177	91	25	1.46	A-C
Joint filler	2	18	72	29	6	2.81	A-C
Sericite	3	9	27	17	6	2.88	C

4.2. An statistical analysis

Plastic failure depends on yielding to applied stress at a given consolidation state. The mathematical formalism needed to describe plasticity has to include the dependence on the history of force applications. To explore a potential μ relation in the quasi-static regime, we begin by data mining the four major parameters obtained from the MCP as independent variables:

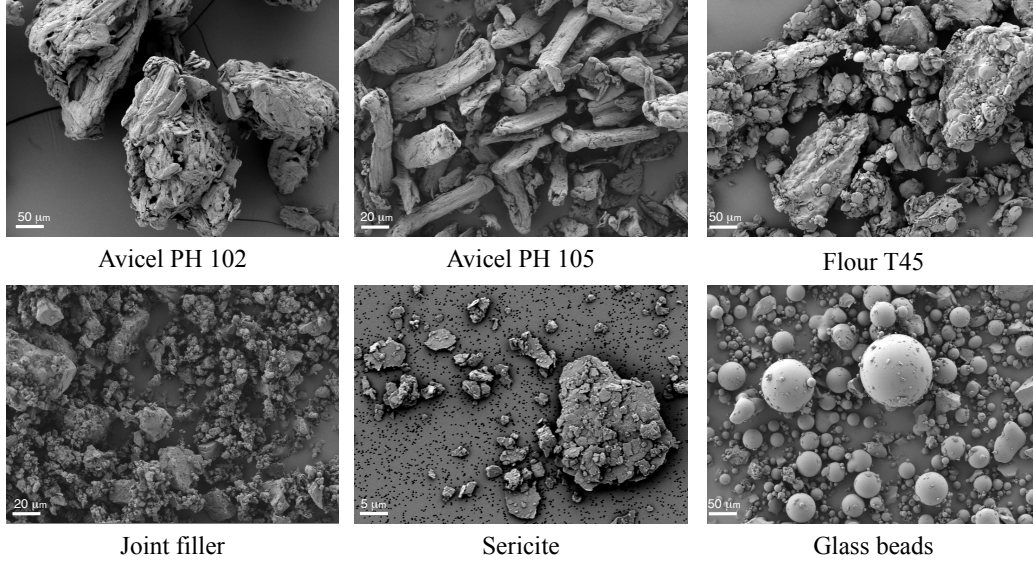


Figure 2: SEM micrographs of samples dataset 3.

μ , σ_p [kPa], c [kPa], and ff . The first dataset contains 287 data points in a four-dimensional space visualized in a scatter plot matrix on a logarithmic scale in figure 3.A. We infer a similarity between most scatterplots from the figure in which the points are evenly distributed into straight-line groups with different inclinations, except for the μ . For a proper interpretation, using MATLAB, we realised a Principal Component Analysis (PCA) showing that μ does not contribute to data variance (noise level). Surprisingly, and unlike the idea which is commonly proposed, the coefficient of friction does not seem to describe the flow. Neglecting μ , our MCP dataset, on a logarithmic scale, can be well evaluated using multiple linear regression. We have a power-law relation between the flow factor and the cohesion in which the law's exponential is close to -1, and the constant depends on the pre-consolidation stress:

$$ff = (a_0 \sigma_p^{a_1}) c^{a_2} \quad (3)$$

where $a_0 = 0.589 \pm 0.007$, $a_1 = 0.934 \pm 0.006$ and $a_2 = -0.948 \pm 0.006$. Fit $R^2 = 0.991$, adjusted $R^2 = 0.991$, p-value ~ 0 .

The PCA also reveals that our system can be characterised by simply two

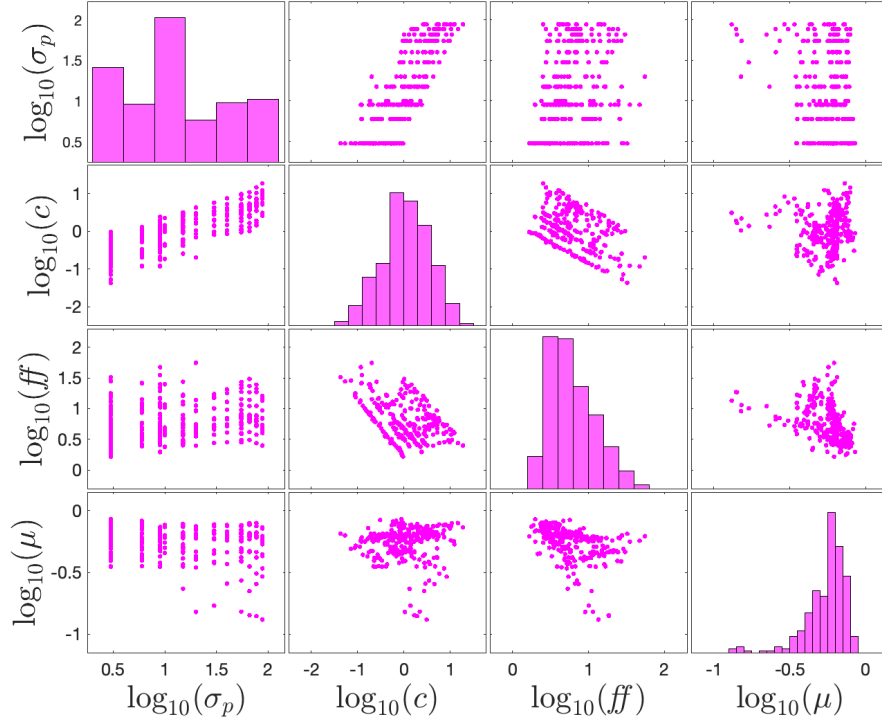


Figure 3: Scatter plot matrix of our dataset composed of 287 data points in a logarithmic scale with pre-consolidation stresses varying between 3 and 88 kPa. The images are the scatter plots of the data in two dimensions at a time, and the diagonal is the histogram of each variable.

variables. As adhesive interactions act similarly to confining stress and tend to prevent dilution during flow, we can define a second dimensionless number that we call the adhesion number $C = c/\sigma_p$ which compares the adhesive interactions between grains with the confining normal stress. The rheology in the quasi-static regime is then described only by two dimensionless variables C and ff , we have

$$ff = (0.58 \pm 0.01) C^{-0.94 \pm 0.01} \quad (4)$$

with the adhesive interactions c varying from 0.04 to 18.86 kPa and ff from 2 to 55.

Despite the diversity of the samples, the environmental conditions and

conditioning all data points fall on a line, showing a power-law relation between the flow factor and the adhesion number, in which the law’s exponential is close to -1 (Fig. 4). Although the residuals seem to be equally spread over the space we found a more significant variability for the cohesion c in free-flowing powders and at high pre-consolidation stresses related to the stiffness and elasticity of the samples (not shown here).

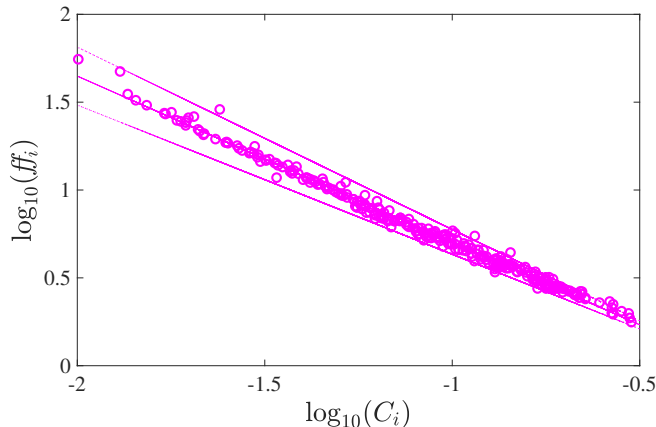


Figure 4: Statistical analysis dataset 1 composed of 287 data points on a logarithmic scale with σ_p varying between 3 and 88 kPa. The solid line represents the model (Eq. 4) and the dashed lines correspond to the 90% confident interval of the fit, $R^2=0.991$, adjusted $R^2=0.991$, p-value 0. Each data point is an average value of at least 3 measurements.

4.3. A phenomenological model from complex granular materials

To test the generality of our model and avoid device-related conclusions, we study the data available in the literature [26, 28, 27, 9, 6, 29, 17]. This second dataset contains 433 data points (including dataset 1) in a two-dimensional space (Fig. 5). These new data points were obtained using the FT4 rheometer [26, 28, 6, 17], the Brookfield powder flow tester (PFT) [9], the Jenike shear cell tester [9], the Schulze ring shear tester [9, 27] or the HT-ASC [29]. Despite the diversity of the samples, the different shear testers and operators and the many environmental conditions and conditioning, the multiple linear regression analysis confirms a power-law relation. These surprisingly accurate experimental results ($R^2 \sim 0.99$, p-value $\sim 0!$) from the MCP continuum approach indicate that adhesive interactions are the primary parameter controlling flow in the quasi-static regime, we have

$$ff = (0.60 \pm 0.01) C^{-0.94 \pm 0.01} \quad (5)$$

with ff calculated by data mining over 120 industrial granular materials with varying degrees of cohesion: c varying from 0.01 to 18.86 kPa) and ff from 1 to 175.

As shown in figure 5, most data gathers in a prediction interval of 90%, with two data subgroups [29, 28] in the 80-90% interval. This is unusual since a phenomenological law obtained from a large device-independent dataset of industrial granular materials like the one presented herein has never, to our knowledge, been identified previously.

The two out-of-range data points were obtained for calcium carbonate using the PFT and are part of the same study [9]. The lack of fit is attributed to the highly cohesive nature of calcium carbonate and the low consolidation used during the test. The sample probably does not yield under the Mohr-Coulomb criterion, which is expected for highly cohesive samples. In Salehi et al., 2017, the authors use low pre-consolidation stresses and compare different ring shear devices, increasing our model's global standard error for prediction (Fig. 5). From their work, we also distinguish calcium lactate (free-flowing sample) with 37% of the data over 20% error attributed to a bi-modal error distribution related to the differences among testers.

Data analysis based on the shear-to-failure device showed a higher standard deviation for the PFT followed by the Schulze shear tester. It should be noted that most of the data were obtained using the FT4 device.

A similar statistical analysis was performed by Gaume and co-workers using the Discrete Element Method to simulate Couette cell experiences for granular samples composed of rigid spherical grains of an average diameter of $d=4.7$ mm [3]. The authors proposed a unique relationship between μ , I and Θ in the power-law form $\Theta = 0.59I^{\beta(\mu)}$ which was validated in the inertial regime. Despite the similarities (pre-factor and law power relationship), in our case adhesive interactions deeply affect the packing properties as well as the flow, prevailing over the friction and profoundly differentiating the description of the flow in the quasi-static regime.

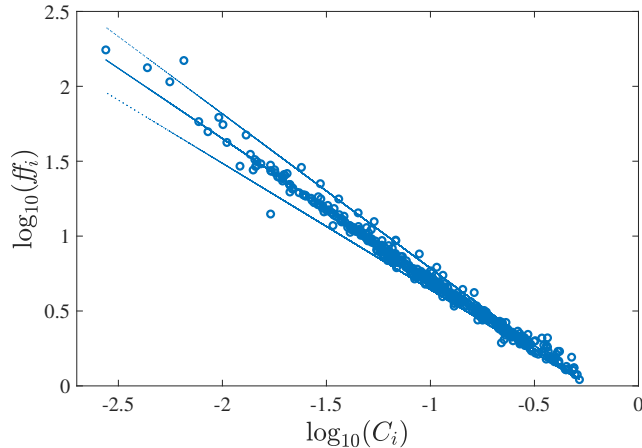


Figure 5: Statistical analysis dataset 2 in a two-dimensional space (C and ff) composed of 433 data points on a logarithmic scale with σ_p varying between 0.69 and 88 kPa. The solid line Parity plot of predicted values of the flow factor $ff = 0.60 \cdot C^{-0.94}$ using multilinear regression analysis. The solid line represents the model, and the dashed lines correspond to the 90% confident interval of the fit, $R^2=0.987$, adjusted $R^2=0.987$, p-value=0.

4.4. Neither the friction nor the packing fraction describe the granular rheology in the quasi-static regime.

As granular rheology in the dense flow regime is often described by the coefficient of friction and volume fraction, we have studied the influence of ϕ in the yield criteria. To study ϕ from shear test measurements, we selected six powders as varied as flour and glass beads (see Fig. 2 and Table 1). A third dataset of 146 data points with σ_p between 3 and 88 kPa was studied. The ϕ values were estimated from raw data and found to be σ_p dependent. The packing fraction remained constant during the analysis (less than 3% variation) despite the changes in consolidation during shearing (Fig. 1).

Figure 6.A shows that the packing fraction increases with the σ_p stress until reaching a plateau. Conversely to what is commonly assumed [3, 30, 4, 2], our results show that the friction coefficient is not always independent of the pre-consolidation stress in the quasi-static regime (Fig. 6.B). For two samples - flour and sericite - we have a diminution of μ with σ_p , explained probably by their lowest elastic modulus. Indeed, these results, and many other of our experimental data (work in progress), show that grains' mechanical properties such as the elastic moduli, play a major role in the bulk friction. This

demonstrates the independence between both parameters when the nature of the grain is taken into account.

The water content of the samples (conditioned at 30% or 60% RH) can influence or not their flow behaviour. Nevertheless, as for datasets 1 and 2, the moisture content is reflected in both C and ff parameters, and they continue to relate accordingly to Eq. 4 and 5.

The PCA from ff , C , μ , and ϕ parameters in logarithmic scale, shows that neither ϕ or μ explains the data. No one-to-one relationships between ϕ or μ and c or ff in the quasi-static regime has been found. For example, samples with comparable volume fractions, such as Avicel PH 102 and Sericite ($\phi \approx 0.25$), have drastically different flow behaviour; although Avicel PH 102 flows easily while Sericite is cohesive and prone to agglomeration (Fig. 6.A).

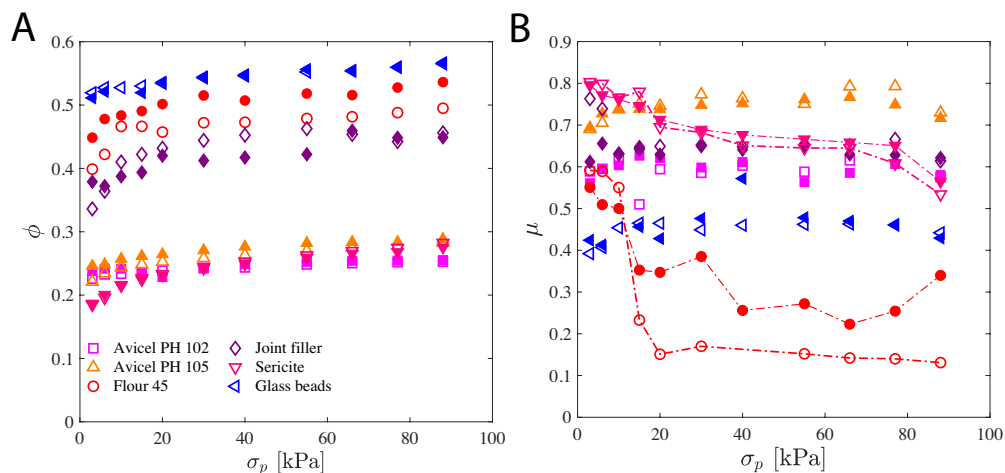


Figure 6: Relations between $\phi(\sigma_p)$ (A) and $\mu(\sigma_p)$ (B) bulk parameters. The dataset is composed of 146 data points obtained from 6 powder samples: Avicel PH102 (\square), Avicel PH 105 (\triangle), Flour T45 (\circ), Joint filler (\diamond), Sericite (∇) and Glass beads (\triangleleft) (see Fig. 2 – online repository). Each sample was conditioned in an air humidity-controlled environment at 30% and 60% RH. The data obtained from samples conditioned at 30% RH is represented by filled marks and unfilled marks when conditioned at 60% RH. The σ_p values are 3, 6, 10, 15, 20, 30, 40, 55, 66, 77 and 88 kPa. Each data point is an average value of at least 3 measurements.

It is observed that the greatest normal stress applied to the bulk defines the solid state. As previously postulated [3], these findings suggest that fluctuations in the quasi-static regime are maintained by boundary conditions imposed at the boundaries of the quasi-static zones.

4.5. *An attempt to unified flow behaviour and interparticle adhesive interactions.*

Powder technology groups have recently attempted to predict granular flow in the quasi-static regime by relating ff to the population-dependent granular Bond number, Bo_G .

$$ff = \alpha(Bo_G)^{-\beta} \quad (6)$$

where α and β are either poorly characterised or assumed to be material and equipment dependent.

Unlike the well-known granular Bond number, defined as the ratio of attractive interparticle force to gravity force, $Bo_g = N_c/W_g$, the Bo_G can account for complex size distributions [31, 17] and may be employed in multi-component mixtures [32]

$$Bo_G = \left(\sum_{i=1}^N \sum_{j=1}^N \frac{w_{ij}}{Bo_{g,ij}} \right)^{-1} \quad (7)$$

where w is a weighted harmonic means of the Bo_g for every possible interaction between different size grains or materials within a blend, i and j represent these variables.

From Eq. 6 various α and β values have been found, each of which is dependent on the σ_p supplied to the bulk during the shear test, such as: for pharmaceutical excipients ($\alpha = 15.7$ and $\beta = 0.27$ with $\sigma_p = 1$ kPa [32]), pharmaceuticals ($\alpha = 14.8$ and $\beta = 0.28$ with $\sigma_p = 1$ kPa [31]) and raw ceramic and grounded powders ($\alpha = 89.5$ and $\beta = 0.54$ with $\sigma_p = 9$ kPa [17]). In the following, as Eq. 5 has a larger spectrum than the equations proposed in these previous works, it will be used to determine α and β values. And, in order to seek a physical meaning of α and β constants, the basic theory is declined.

Rumpf [33] described interparticle interaction and bonding by relating the cohesive stress, σ_c , to the force per contact N_c and the microstructure, described by ϕ and the coordination number, Z , assuming there is an isotropic granular material, an isotropic contact network and that all contacts break at the shear threshold in two dimensions

$$\sigma_c \approx \frac{\mu \phi Z N_c}{\pi d^2} \quad (8)$$

Given that macroscopic adhesive stress from the Coulomb criterion represents the yield shear stress at zero confining stress, it is reasonable to assume that $\sigma_c \approx c$. Theoretical predictions based on Rumpf's equation show a linear relationship between c , and the interparticle adhesive force strength [34, 35, 36, 37, 38, 17, 2]. The bulk cohesiveness may be expressed using the Bond number as

$$c \approx \frac{\mu \phi Z W_g B o_g}{\pi d^2} = \frac{\rho_s g \mu \phi Z d B o_g}{6} \quad (9)$$

From our model (Eq. 5), we propose the following form for Eq. 6:

$$ff = 2.72 \left(\frac{\rho_g \mu \phi Z}{\sigma_p^{0.97}} B o_G \right)^{-0.96} \quad (10)$$

The predictability of Eq. 10 is too sensitive to $B o_G$ to determine flow behaviour, and N_c remains quite complicated to quantify. Thus using our samples or those from the literature [32, 31, 17], the predictability can be statistically significant or quite disappointing. Also, it should be noted that C experimental values are much smaller than those predicted by Rumpf's formula (Eq. 8) as previously reported and attributed to the anisotropy in the contact network [36, 2].

Mathematically, the main problem in Eq. 10 seems to be the exponential value. Giraud et al. [17] proposed to take into account the size distributed properties using Rosin-Rammler law to evaluate Eq. 5 using $B o_G \propto d_g^{-2.6}$. Thus, Eq. 10 gives

$$ff = 2.72 \left(\frac{d \rho_g \mu \phi Z}{\sigma_p^{0.97}} \right)^{-0.96} B o_G^{-0.59} \quad (11)$$

We did not successfully validate the relationship between interparticle forces and macroscopic flow behaviour using Eq. 11, which resulted in being less accurate than Eq. 10. The difficulties in constructing Eq. 6 is that interparticle interactions depend on the particle size and surface topography [25, 39, 40] and on the local radius of curvature at contact. Particle size has been the subject of much attention since this is the simplest and most commonly used approach to characterise grains. Nonetheless, we found no linear relationship between particle size and bulk adhesive forces for small particles

(Sauter diameter, $d_{3,2} \leq 200\mu\text{m}$). This means that the other anisotropies in the contact (much more challenging to quantify) are responsible for the deviation from Rump’s model.

To finally achieve the breakthrough needed in granular flow understanding, contact mechanics theories and their experimental assessment must be improved in order to establish the relationship between interparticle forces and bulk behaviour.

The relation between interparticle adhesion and bulk adhesion from the yield locus-MCP is worthy of further investigation and validation. The highly predictive capacity and the simplicity of our model (Eq. 5) could be of great use in industrial applications involving formulation, processing, storage and transport. We foresee a power-law model relating both macroscopic bulk properties and microscopic grain properties to describe granular flows in the quasi-static-regime $ff = 0.60 C^{-0.94} \approx \alpha(Bo_G)^\beta$, with α and β parameters to be properly estimated. Careful attention is required because, while the flow factor proposed by Jenike is adapted to silo-hopper engineering where the flow field is steady from an Eulerian perspective, it will not describe the flow behaviour of bulk solids by simply pouring them. The whole flow dynamic of granular matter, from quasi-static to gaseous regime, is beyond the scope of this work.

5. Conclusion

In this research, a phenomenological law from the Mohr-Coulomb plastic failure theory has been identified by data mining over 130 industrial granular materials with varying degrees of cohesion. This is unusual since a statistical analysis obtained from a large dataset of industrial granular materials, device-independent like the one presented herein, has, to our knowledge, never previously been identified.

Several studies and industrial know-how have proven the importance of grains’ intrinsic/extrinsic properties when describing granular flows with most of them related to grain size and friction. Contrary to what is commonly perceived, we found that granular rheology in the quasi-static regime is governed and defined by adhesive interactions, via the Adhesion number $C = c/\sigma_p$, rather than friction. Our results have been shown to be remarkably accurate ($R^2 \sim 0.99$, p-value ~ 0). Furthermore, due to particle mechanical properties, the traditional assertion, $\mu \sim \text{cte}$, does not apply to complex

grains. The authors would not argue that size, packing fraction, and friction are irrelevant elements in granular flow, rather that C encompasses them in the quasi-static regime. One of the key reasons for the problems in linking motion from the quasi-static to the dense flow regime might be this.

The MC theory is based on the assumption that granular materials may be considered to be rigid-plastic continuous media. Our findings suggest that fluctuations in the quasi-static regime are maintained by boundary conditions imposed at the boundaries of the quasi-static zones. From a general perspective, granular materials are not continuous and mechanical integrity is not granted, these are the limitations of the Mohr-Coulomb plasticity (MCP) in describing the flowability of granular materials. Nevertheless, in the quasi-static regime it remains the most accurate and widely used model for granular materials in engineering and powder technology. Some other plastic flow rules have been proposed giving rise to several size-dependent continuum models [41, 42]. Still, none of them sharply describe the flow behaviour of day-to-day granular materials such as those used in our work (flour, cement, mica, etc.).

Fully understanding the concept of flowability from solid-like to fluid-like and then to gas-like remains to be achieved. We have indicated the benefits of using experimental data like those presented herein. In our view, the lack of a theoretical basis using this kind of information has undermined overall understanding and fostered the rather unjustified idea that frontier science cannot describe granular flows for industrial applications. Much more effort is required to combine cross-disciplinary scientific and technological skills to relate fundamental physics to the flow behaviour of more “complicated” granular materials, such as powders. This work is our “first brick in the wall” contribution as a concerted effort from research and industry.

All data presented in this article is available openly in DOREL repository from the Université de Lorraine, <https://doi.org/10.12763/EKRLDI>.

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