



HAL
open science

Grain size distribution does not affect the residual shear strength of granular materials: An experimental proof

Oscar Polanía, Miguel Cabrera, Mathieu Renouf, Emilien Azéma, Nicolas Estrada

► To cite this version:

Oscar Polanía, Miguel Cabrera, Mathieu Renouf, Emilien Azéma, Nicolas Estrada. Grain size distribution does not affect the residual shear strength of granular materials: An experimental proof. *Physical Review E*, 2023, 107 (5), pp.L052901. 10.1103/PhysRevE.107.L052901 . hal-04097310

HAL Id: hal-04097310






<https://hal.science/hal-04097310v1>

Submitted on 15 May 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Grain size distribution does not affect the residual shear strength of granular materials: An experimental proof

Oscar Polanía ^{1,2,*}, Miguel Cabrera ^{3,†}, Mathieu Renouf ^{2,‡}, Emilien Azéma ^{2,4,§} and Nicolas Estrada ^{1,||}

¹Department of Civil and Environmental Engineering, Universidad de los Andes, Bogotá, Colombia

²LMGC, Université de Montpellier, CNRS, Montpellier, France

³Department of Geoscience & Engineering, TU Delft, Delft, The Netherlands

⁴Institut Universitaire de France (IUF), Paris, France



(Received 13 March 2023; accepted 20 April 2023; published 12 May 2023)

Granular materials are used in several fields and in a wide variety of processes. An important feature of these materials is the diversity of grain sizes, commonly referred to as polydispersity. When granular materials are sheared, they exhibit a predominant small elastic range. Then, the material yields, with or without a peak shear strength depending on the initial density. Finally, the material reaches a stationary state, in which it deforms at a constant shear stress, which can be linked to the residual friction angle ϕ_r . However, the role of polydispersity on the shear strength of granular materials is still a matter of debate. In particular, a series of investigations have proved, using numerical simulations, that ϕ_r is independent of polydispersity. This counterintuitive observation remains elusive to experimentalists, and especially for some technical communities that use ϕ_r as a design parameter (e.g., the soil mechanics community). In this Letter, we studied experimentally the effects of polydispersity on ϕ_r . In order to do so, we built samples of ceramic beads and then sheared these samples in a triaxial apparatus. We varied polydispersity, building monodisperse, bidisperse, and polydisperse granular samples; this allowed us to study the effects of grain size, size span, and grain size distribution on ϕ_r . We find that ϕ_r is indeed independent of polydispersity, confirming the previous findings achieved through numerical simulations. Our work fairly closes the gap of knowledge between experiments and simulations.

DOI: [10.1103/PhysRevE.107.L052901](https://doi.org/10.1103/PhysRevE.107.L052901)

Granular materials are found in several fields, such as the construction, pharmaceutical, and food industries, involving a wide variety of processes such as compaction, mixing, storage, and fragmentation, among others. When these materials are sheared, they exhibit a small range of deformation in which the material behaves elastically and deformations are reversible. Beyond this elastic regime, plasticity develops and irreversible deformations start to accumulate. As both volumetric and distortional deformations increase, the shear strength increases too, up to a limit that is highly dependent on the initial density of the arrangement. Afterwards, a stationary state is reached, in which shear deformations continue to accumulate while volume and shear strength fluctuate around approximately constant values. This stationary shear strength is termed the *residual strength* of the material, and it can be described by the residual angle of internal friction ϕ_r . The residual strength is of primary importance in the design of processes involving large deformations of granular materials or in the characterization of granular flows [1,2].

The packing efficiency of a granular system can be quantified by means of the packing fraction ν ,

$$\nu = \frac{V_g}{V}, \quad (1)$$

where V_g is the volume occupied by the grains and V is the total volume. For monosized spheres, the reference values are random loose packing $\nu_l \simeq 0.55$ and random close packing $\nu_c \simeq 0.64$ [3]. In many cases, granular materials have grains of different sizes, and thus are referred to as polydisperse. Sedimentary soils, concrete aggregates, or coke and ore grains needed for the production of steel are all examples of polydisperse granular materials. Polydisperse materials can have larger packing fractions than monodisperse systems, because small grains partially fill the voids between larger grains.

An important variable that controls ν is the grain size distribution (GSD). During the last decade, several investigations conducted by means of the discrete element method (DEM) have suggested that ν is maximized for GSDs in which all size classes occupy the same volume (i.e., for a uniform distribution by volume fraction) [4,5]. This disagrees with an important finding dating back to the early 20th century, where Fuller and Thomson found that the densest arrangement is obtained when the GSD has a cumulative volume fraction CVF that follows a power law of the form

$$\text{CVF} = \left(\frac{d}{d_{\max}} \right)^t, \quad (2)$$

*os.polania@uniandes.edu.co; oscar.polania@umontpellier.fr

†M.A.Cabrera@tudelft.nl

‡mathieu.renouf@umontpellier.fr

§emilien.azema@umontpellier.fr

||n.estrada22@uniandes.edu.co

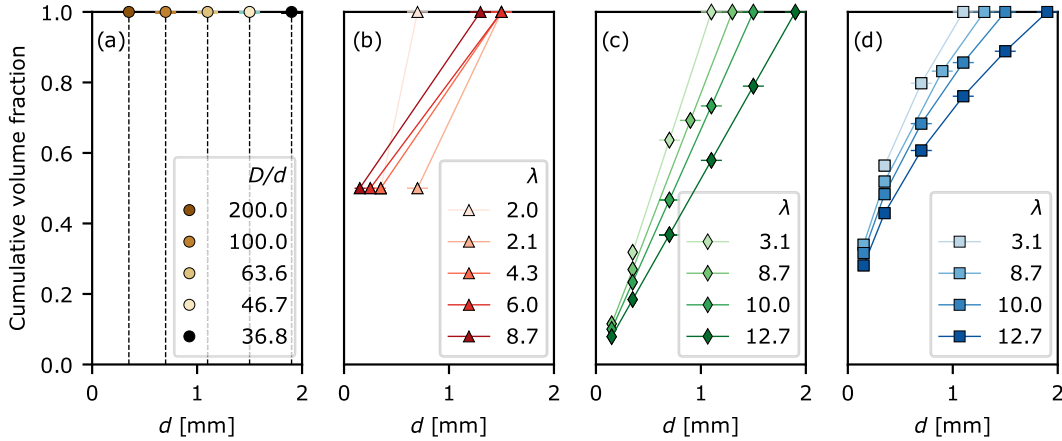


FIG. 1. Cumulative volume fraction CVF as a function of grain diameter d , for the four types of grain size distributions explored in this Letter: (a) Monodisperse samples, (b) bidisperse samples, (c) polydisperse samples with a uniform distributions by volume fraction, and (d) polydisperse samples with a distribution following $\text{CVF} = (d/d_{\max})^{0.5}$. Symbols represent the average and horizontal bars represent the range in size reported by the supplier Sigmund Lindner GmbH. $\lambda = d_{\max}/d_{\min}$ is the size ratio between the maximum d_{\max} and minimum grain diameter d_{\min} . For monodisperse samples (a), D/d is the system-to-grain size ratio, with $D = 70$ mm being the sample's diameter.

where d is the grain diameter, d_{\max} is the maximum grain diameter, and the exponent $\iota \simeq 0.5$ [6]. In this form, the uniform distribution by volume fraction corresponds to $\iota = 1$. More recently, also by means of DEM simulations, several numerical studies have systematically explored the effect of ι on ν , confirming that the densest and better connected arrangement is obtained for $\iota \simeq 0.5$, as it was proposed by Fuller and Thompson [7–9].

It is well known that the shear strength of granular materials depends on material properties such as the grains' shape or the friction coefficient between grains. Polydispersity, as a fundamental feature of granular materials, has also been explored in order to identify its influence on the shear strength. However, this relationship remains a matter of debate, especially in the soil mechanics community, even if one restricts the question to the residual (i.e., the simplest) angle of internal friction ϕ_r . On the one side, experimental studies have shown that ϕ_r decreases with the level of polydispersity [10–12], which can be quantified by means of the grain size span $\lambda = d_{\max}/d_{\min}$, while others have observed that ϕ_r increases with λ [13–17]. In the framework of critical state soil mechanics, some studies have explored the effects of the GSD and shown that the critical state of soils is independent of size [18] and level of polydispersity [19,20]. There is thus no agreement on this effect in the soil mechanics community. On the other side, a good number of DEM numerical studies have shown that ϕ_r is independent of both λ and ι [7,21–27]. These studies have also allowed explaining this independence as a compensation mechanism between the different anisotropies that can be defined in the contact and the force networks [21–23,25,26,28,29]. There is thus agreement on the independence of ϕ_r and polydispersity in the granular-DEM community.

The aim of this Letter is to conduct a set of systematic and controlled experiments in order to confirm if ϕ_r is independent of polydispersity, described in terms of grain size (d), size span (λ), and grain size distribution (λ and ι). To the authors' knowledge, a systematic experimental campaign aiming to

explore the link between polydispersity and the residual shear strength has not yet been presented.

In order to do this, we performed a set of drained (i.e., dry) triaxial tests with spherical ceramic beads of different sizes. Using these grains, we built 18 samples with different GSDs: monodisperse, bidisperse, polydisperse with a uniform distribution by volume fraction, and polydisperse with a Fuller and Thompson distribution. Our samples had size spans $\lambda \in [1.0\text{--}12.7]$ (see Fig. 1). We used this type of grains in order to exclude the effects of angularity, elongation, and contact friction, which allowed us to observe solely the effects of the GSD.

The triaxial test is widely used in soil and rock mechanics. In this test, the cylindrical granular samples are placed inside a flexible membrane, which allows for applying a lateral confining pressure σ_3 that is kept constant throughout the experiment. Then, the samples are compressed axially at a constant rate $\dot{\epsilon}_1$, inducing a vertical stress σ_1 . In our experimental campaign, we used samples of height $H = 140$ mm and diameter $D = 70$ mm. The vertical strain ϵ_1 was computed as $\Delta H/H$. An advantage of this test is that the shear band develops naturally inside the sample and not on preimposed failure planes. In the triaxial test, although the shear direction is not controlled and the shear stress cannot be measured directly, a normalized shear stress q/p can be computed from the mean $p = (\sigma_1 + 2\sigma_3)/3$ and deviatoric $q = \sigma_1 - \sigma_3$ stresses. According to the Mohr-Coulomb model, the friction coefficient is $\sin \phi = 3q/(6p - q)$.

For monodisperse samples, we varied d between 0.35 and 1.9 mm. In bidisperse samples, the mass of each species was 50% of the total mass. For polydisperse samples, we used samples that had from three to six grain sizes, with which we built two different GSDs: uniform distributions by volume fraction, and Fuller and Thompson distributions (i.e., GSDs with $\iota = 1$ and $\simeq 0.5$, respectively). The cumulative volume fraction CVF of the 18 samples used in this work is shown in Fig. 1. We used ceramic beads with density $\rho = 6.05$ g/cm³ of the type ZY produced by Sigmund Lindner GmbH

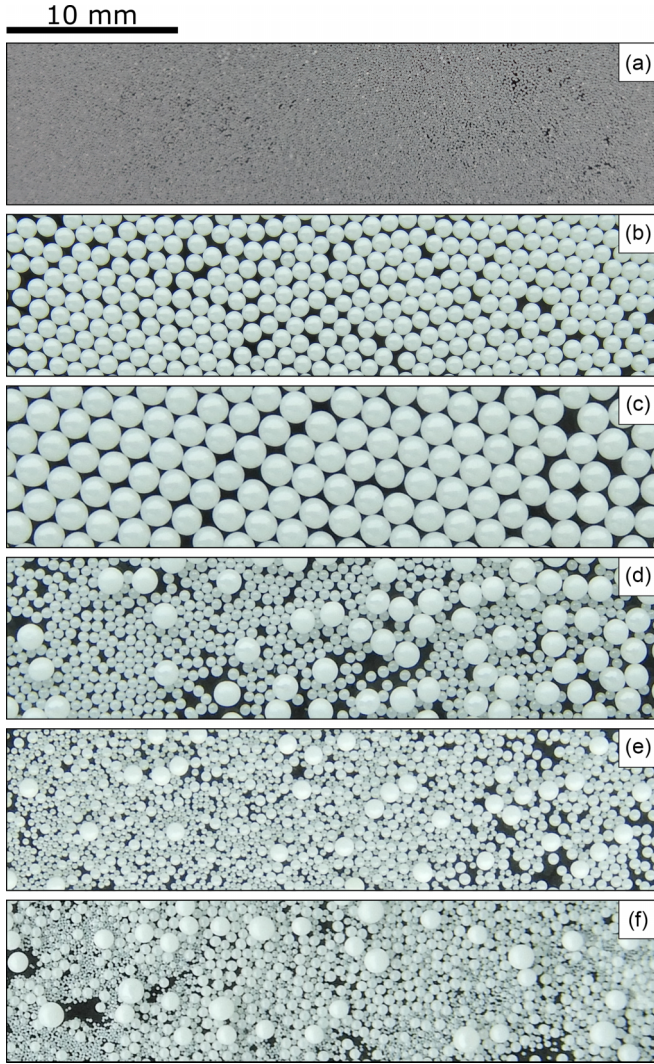


FIG. 2. Close-up scale views of some of the tested granular systems. (a)–(c) Grains with diameters $d = 0.15 \pm 0.05$ mm, $d = 1.1 \pm 0.1$ mm, and $d = 1.9 \pm 0.1$ mm, respectively, (d) bidisperse sample with $\lambda = 2.1$, and (e) and (f) polydisperse samples with $\lambda = 3.1$ and 10, respectively. A 10 mm scale is shown in the upper-left-hand corner.

(see Fig. 2). We acknowledge that many granular systems, especially those involved in geophysical processes [30,31], might have larger values of λ than the ones we explored, but we consider that the size span used is representative to study the residual resistance of polydisperse granular materials.

Sample preparation was started by hand mixing the grains in a large container until having an apparently homogeneous mixture. To diminish segregation during sample preparation, we gently poured the mix with a long spoon inside the cylinder. The cylinder is filled in three layers, densifying each layer by tamping and shuddering up to a flat surface. The initial packing fraction ν_0 rapidly increased with λ , and it reached a plateau for $\lambda > 4$ at $\nu_0 \approx 0.71$. The maximum ν_0 occurred for $\lambda = 12$ and $\iota = 0.5$. Excluding monodisperse systems and bidisperse with $\lambda = 2$, our samples had initial packing fractions that were larger than that of the random close packing ν_c (see Fig. 3).

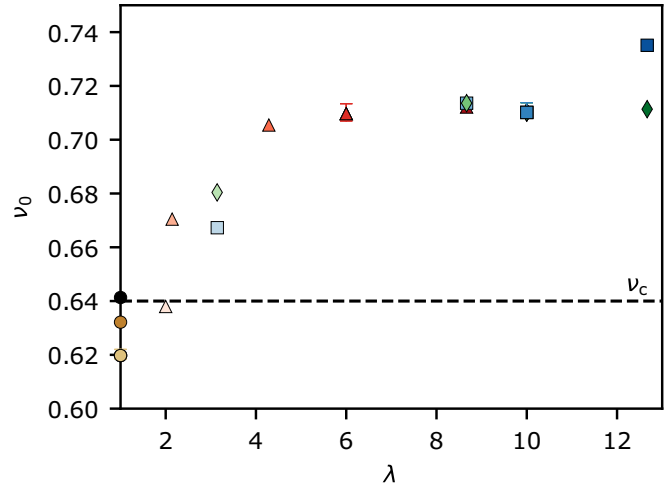


FIG. 3. Initial packing fraction ν_0 as a function of the size span λ . The known limit of the random close packing for monodisperse spheres, $\nu_c = 0.64$, is shown by the dashed line [3]. Error bars are the minimum and maximum values between sample repetitions. Monodisperse samples (\circ); bidisperse samples (\triangle), polydisperse samples with uniform distributions by volume fraction (i.e., with exponent $\iota = 1$) (\diamond), and polydisperse samples with Fuller and Thompson distributions (i.e., with exponent $\iota = 0.5$) (\square). The symbols' colors match those in Fig. 1.

The test inertial level was computed with the inertial number $I = \dot{\epsilon}_1 \bar{d} / \sqrt{\sigma_3 / \rho_p}$, where \bar{d} could be either d_{\max} or d_{\min} , and $\dot{\epsilon}_1$ is the vertical strain rate. Tests were conducted at a confinement pressure $\sigma_3 = [100, 200]$ kPa and the vertical strain rate is set to $\dot{\epsilon}_1 = 0.0001/\text{s}$, resulting, in all cases, in quasi-static experiments with $I < 10^{-4}$. Samples were deformed up to $\epsilon_1 = 0.2$ and the residual strength was considered at $\epsilon_1 \in [0.15, 0.2]$. Finally, we sieved the samples and confirmed that no modifications of the GSD (e.g., grain crushing) had occurred at the end of the test. In order to quantify variability, we conducted three repetitions for one GSD of each set [i.e., three repetitions of the monodisperse sample with $d = 1.1$ mm, the bidisperse sample with $\lambda = 6$, the polydisperse sample with $(\lambda, \iota) = (10, 1)$, and the polydisperse sample with $(\lambda, \iota) = (10, 0.5)$].

Figure 4 shows the evolution of the normalized shear stress q/p for the four GSDs investigated in this work. For monodisperse systems, we found that the shear strength of samples with $d = 1.9$ mm is affected by grain size effects (i.e., for a system-to-grain size ratio $D/d \lesssim 37$). This holds true for both values of σ_3 . For samples with $D/d \gtrsim 46$, q/p converges to a common value [see Fig. 4(a)]. This result shows that, provided that the sample is sufficiently large compared to the grains' size, the shear strength is independent of the grain size, which is in agreement with previous experimental results [18,32]. After identifying the grain size effect for samples with $D/d \lesssim 37$, we decided to not include grains with $d > 1.9$ mm in bidisperse samples. Interestingly, for all values of the size span λ in bidisperse experiments, q/p also tends to a common value. This result shows that the residual strength is not only independent of grain size, but it is also independent of the size span [see Fig. 4(b)]. Polydisperse samples with exponents $\iota = 1$ and 0.5 show, with greater clarity, that q/p is in

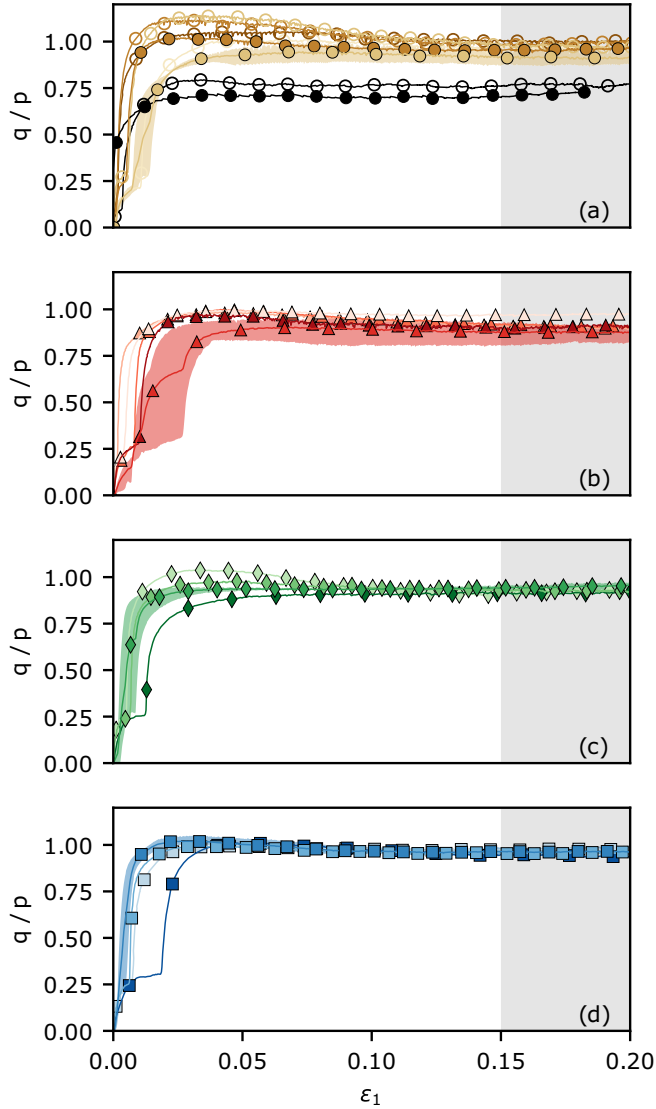


FIG. 4. Evolution of the normalized shear stress q/p as a function of the vertical strain ϵ_1 for (a) monodisperse samples, (b) bidisperse samples, (c) polydisperse samples with uniform distributions by volume fraction (i.e., with exponent $\nu = 1$), and (d) polydisperse samples with Fuller and Thompson distributions (i.e., with exponent $\nu = 0.5$). Shaded areas represent the variability between repetitions with envelopes indicating the minimum and maximum values. The gray shaded region between $\epsilon_1 \in [0.15, 0.2]$ is the region where the residual shear strength is computed. In (a), open (\circ) and solid markers (\bullet) correspond to experiments with $\sigma_3 = 100$ and 200 kPa, respectively. The symbols' shapes and colors match those in Fig. 1.

fact independent of both λ and ν [see Figs. 4(c) and 4(d)]. This result confirms experimentally that the residual shear strength is independent of the grain size distribution, being in agreement with previous results obtained by means of DEM simulations [7,21–27].

Now, let us compare the residual shear strength of all GSDs analyzed in this work. Figure 5 shows the residual friction angle ϕ_r for all samples as a function of the size span λ . It can be seen that all values collapse around a common value of $\phi_r \simeq 24$. Bidisperse samples exhibit higher variability, even

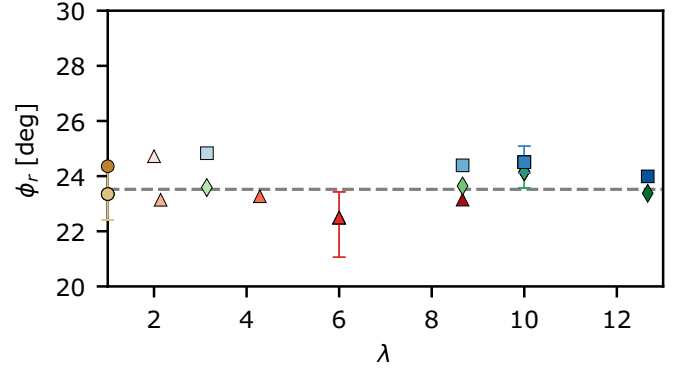


FIG. 5. Residual friction angle ϕ_r as a function of the size span λ . The dashed line shows the mean value for all experiments. Vertical bars indicate the maximum and minimum values between repetitions. Monodisperse samples (\circ), bidisperse samples (\triangle), polydisperse samples with uniform distributions by volume fraction (i.e., with exponent $\nu = 1$) (\diamond), and polydisperse samples with Fuller and Thompson distributions (i.e., with exponent $\nu = 0.5$) (\square). The symbols' colors match those in Fig. 1.

though ϕ_r is close to that of monodisperse and polydisperse samples. This result is in disagreement with the one presented in Ref. [33], where a strong contrast between bidisperse and polydisperse systems was found. It can also be seen that polydisperse samples with $\nu = 0.5$ have a slightly larger ϕ_r than the rest of GSDs. In order to understand this small difference, one must take into account that these are also the samples with the highest initial density and that, by design, the triaxial test is incapable of fully reaching a steady state. For example, the triaxial tests conducted in this work reach a maximum $\epsilon_1 = 0.2$ before the sample collapse. It is thus probable that the shear strength of these samples is still marginally affected by the sample' initial density.

In summary, we conducted an experimental campaign of dry triaxial tests, varying the grains' size, size span, and size distribution of samples composed of ceramic beads. Our experiments allowed us to study the effects that these three features have on the residual shear strength of granular materials, since all samples were built with spherical grains, with the same contact friction coefficient, and at a level of confinement that was low enough in order to prevent crushing. Overall, our results confirm that the residual shear strength of granular materials is independent of the grain size, of the level of polydispersity, and of the CVF shape, as it has been shown numerically [7,21–27] and as it has been suggested in previous experimental works [19,20]. A table with the parameters and results of the experiments that support the conclusions of the present work is included as Supplemental Material [34].

The experimental results presented in this Letter are important because they fairly close the gap of knowledge between experiments and simulations of quasistatic polydisperse systems, confirming that the shear strength of granular materials is independent of the grain size distribution. They also work as a link, connecting results coming from the granular physics and the soil mechanics communities, and directing the discussion of the residual strength towards a common conclusion. The results of this Letter contribute to the knowledge of polydisperse granular materials' behavior that is a matter of

interest in natural and industrial processes. Further studies focusing on the resistance of granular materials might look into other details, such as the link between packing fraction and peak shear strength, or the role of grain shape on the shear response. Contributions of this kind would enrich the understanding of polydisperse granular materials.

The authors are thankful for the support of Julieth Monroy and José Naranjo from the geotechnical laboratory at Universidad de Los Andes. O.P. thanks Miguel David Valencia for his enriching thoughts and stimulating discussions. Moreover, we would like to thank the anonymous referees for their valuable and constructive comments.

- [1] O. Hungr, Dynamics of rapid landslides, in *Progress in Landslide Science*, edited by K. Sassa, H. Fukuoka, F. Wang, and G. Wang (Springer, Berlin, 2007), pp. 47–57.
- [2] G. Mesri and N. Huvaj-Sarihan, Residual shear strength measured by laboratory tests and mobilized in landslides, *J. Geotech. Geoenviron. Eng.* **138**, 585 (2012).
- [3] G. D. Scott, Packing of spheres: Packing of equal spheres, *Nature (London)* **188**, 908 (1960).
- [4] C. Voivret, F. Radjai, J.-Y. Delenne, and M. S. El Youssoufi, Space-filling properties of polydisperse granular media, *Phys. Rev. E* **76**, 021301 (2007).
- [5] P. Mutabaruka, M. Taiebat, R. J.-M. Pellenq, and F. Radjai, Effects of size polydispersity on random close-packed configurations of spherical particles, *Phys. Rev. E* **100**, 042906 (2019).
- [6] W. B. Fuller and S. E. Thompson, The laws of proportioning concrete, *Trans. Am. Soc. Civ. Eng.* **59**, 67 (1907).
- [7] N. Estrada, Effects of grain size distribution on the packing fraction and shear strength of frictionless disk packings, *Phys. Rev. E* **94**, 062903 (2016).
- [8] W. F. Oquendo-Patiño and N. Estrada, Densest arrangement of frictionless polydisperse sphere packings with a power-law grain size distribution, *Granular Matter* **22**, 75 (2020).
- [9] W. F. Oquendo-Patiño and N. Estrada, Finding the grain size distribution that produces the densest arrangement in frictional sphere packings: Revisiting and rediscovering the century-old fuller and thompson distribution, *Phys. Rev. E* **105**, 064901 (2022).
- [10] Y. Liang, T.-C. J. Yeh, J. Wang, M. Liu, and Y. Hao, Effect of particle size distribution on soil-steel interface shear behavior, *Soil Mech. Found. Eng.* **54**, 310 (2017).
- [11] H.-L. Wang, W.-H. Zhou, Z.-Y. Yin, and X.-X. Jie, Effect of grain size distribution of sandy soil on shearing behaviors at soil-structure interface, *J. Mater. Civ. Eng.* **31**, 04019238 (2019).
- [12] L. Carbillet, M. J. Heap, P. Baud, F. B. Wadsworth, and T. Reuschlé, The influence of grain size distribution on mechanical compaction and compaction localization in porous rocks, *J. Geophys. Res.: Solid Earth* **127**, e2022JB025216 (2022).
- [13] T. Kokusho, T. Hara, and R. Hiraoka, Undrained shear strength of granular soils with different particle gradations, *J. Geotech. Geoenviron. Eng.* **130**, 621 (2004).
- [14] A. Simoni and G. T. Houlsby, The direct shear strength and dilatancy of sand–gravel mixtures, *Geotech. Geol. Eng.* **24**, 523 (2006).
- [15] O. Igwe, H. Fukuoka, and K. Sassa, The effect of relative density and confining stress on shear properties of sands with varying grading, *Geotech. Geol. Eng.* **30**, 1207 (2012).
- [16] P. Cao, M. jing Jiang, and Z. jun Ding, Effects of particle size on mechanical behaviors of calcareous sand under triaxial conditions, *Jpn. Geotech. Soc. Spec. Publ.* **8**, 182 (2020).
- [17] Y. Liu, F. Guillard, B. Marks, P. Rognon, and I. Einav, The perpetual shearing of granular soils under low stresses using the stadium shear device, *Open Geomech.* **3**, 2 (2022).
- [18] S. Amirpour Harehdasht, M. Karray, M. N. Hussien, and M. Chekired, Influence of particle size and gradation on the stress-dilatancy behavior of granular materials during drained triaxial compression, *Int. J. Geomech.* **17**, 04017077 (2017).
- [19] G. Li, Y.-J. Liu, C. Dano, and P.-Y. Hicher, Grading-dependent behavior of granular materials: From discrete to continuous modeling, *J. Eng. Mech.* **141**, 04014172 (2015).
- [20] J. Yang and X. Luo, The critical state friction angle of granular materials: does it depend on grading?, *Acta Geotechnica* **13**, 535 (2018).
- [21] C. Voivret, F. Radjai, J.-Y. Delenne, and M. S. El Youssoufi, Multiscale Force Networks in Highly Polydisperse Granular Media, *Phys. Rev. Lett.* **102**, 178001 (2009).
- [22] D.-H. Nguyen, E. Azéma, P. Sornay, and F. Radjai, Effects of shape and size polydispersity on strength properties of granular materials, *Phys. Rev. E* **91**, 032203 (2015).
- [23] D. Cantor, E. Azéma, P. Sornay, and F. Radjai, Rheology and structure of polydisperse three-dimensional packings of spheres, *Phys. Rev. E* **98**, 052910 (2018).
- [24] S. Linero Molina, E. Azema, N. Estrada, S. Fityus, J. Simmons, and A. Lizcano, Impact of grading on steady-state strength, *Géotechnique Lett.* **9**, 328 (2019).
- [25] S. Zhao, J. Zhao, and N. Guo, Universality of internal structure characteristics in granular media under shear, *Phys. Rev. E* **101**, 012906 (2020).
- [26] Y. Zhu, Z. Nie, J. Gong, J. Zou, L. Zhao, and L. Li, An analysis of the effects of the size ratio and fines content on the shear behaviors of binary mixtures using DEM, *Comput. Geotech.* **118**, 103353 (2020).
- [27] M. Cabrera and N. Estrada, Is the grain size distribution a key parameter for explaining the long runout of granular avalanches?, *J. Geophys. Res.: Solid Earth* **126**, e2021JB022589 (2021).
- [28] L. Rothenburg and R. J. Bathurst, Analytical study of induced anisotropy in idealized granular materials, *Géotechnique* **39**, 601 (1989).
- [29] E. Azéma, S. Linero, N. Estrada, and A. Lizcano, Shear strength and microstructure of polydisperse packings: The effect of size span and shape of particle size distribution, *Phys. Rev. E* **96**, 022902 (2017).
- [30] B. Wang, Y. Li, D. Liu, and J. Liu, Debris flow density determined by grain composition, *Landslides* **15**, 1205 (2018).
- [31] L. F. Prada-Sarmiento, M. A. Cabrera, R. Camacho, N. Estrada, and A. M. Ramos-Cañón, The Mocoa event on March 31

- (2017): Analysis of a series of mass movements in a tropical environment of the Andean-Amazonian Piedmont, *Landslides* **16**, 2459 (2019).
- [32] Y. Deng, Y. Yilmaz, A. Gokce, and C. S. Chang, Influence of particle size on the drained shear behavior of a dense fluvial sand, *Acta Geotechnica* **16**, 2071 (2021).
- [33] M. Wackenhut, S. McNamara, and H. Herrmann, Shearing behavior of polydisperse media, *Eur. Phys. J. E* **17**, 237 (2005).
- [34] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevE.107.L052901> for a table with the parameters and results of the experiments that support the conclusions of the present work.