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To cite this version:

Laurent Devriendt, Cendrine Gatumel, Henri Berthiaux. Experimental Evidence of Mixture Segregation by Particle Size Distribution. Particulate Science and Technology, 2013, 31 (6), pp.653-657. 10.1080/02726351.2013.832447 . hal-01649513

HAL Id: hal-01649513

https://hal.science/hal-01649513v1

Submitted on 7 Nov 2019

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Experimental Evidence of Mixture Segregation by Particle Size Distribution

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In this study, we discuss experimental segregation results obtained for two industrial cases, namely, ammonium perchlorate and a polymeric resin. These results show a segregation effect due to particle size distribution rather than particle size itself. We used a heap-pouring device as a tester, for which a visual knowledge of the segregation state was observed. The analysis of segregation is based on various coefficients of variations related to the size fractions or particle size distribution's global characteristics, indicating heterogeneities in the heaps formed. Both cases indicate that wide particle size distributions, as opposed to narrow ones, are limiting segregation risks. This collective, and maybe astonishing, effect is extremely marked for the cases studied, and demonstrates again the mesoscopic nature of granular media.

Keywords: particle segregation, segregation test, particle size distribution, mixing, mixture homogeneity

1. Introduction

Segregation can be viewed as a mechanism leading to a nonrandom degree of uniformity of the different entities that enter in the composition of a product, at the scale at which end-used properties are required. Sources and mechanisms of segregation have been reported and reviewed over many years (Williams 1965, 1976). Particle elutriation, particle bed percolation, trajectories, and vibrations were identified as segregation mechanisms readily to occur during powder chutes, transport, storage, mixing, heaping, and so forth. The effect of segregation on the efficiency of processes involving particle transformations, such as agglomeration, crystallization, grinding, or polymerization (Kim and Choi 2001), is a major industrial issue.

As noted by Tang and Puri (2002), the factors that enhance segregation are extremely diverse and their interactions are far from being known. They also noted the gap between real-case studies and ideal material-based research. Indeed, segregation of particulate solids is a challenging issue in chemical engineering research (see Ottino and Khakar 2001) and of a wide importance in all industrial sectors concerned with the handling of powders: pharmaceuticals, polymers, agro-food, cements, specialty chemicals, explosives, materials, etc. It is undoubtedly a major source of nonconformity of commercial products, with the associated cost usually resulting in a sensible loss of competitiveness for companies. This can be illustrated by some U.S. Food and Drug Administration (FDA) reports, previously commented on by Hussain (2004), pointing out off-specifications rates rising to 50% in several factories.

Segregation tests have been developed at both academic and industrial levels by placing the powders in a critical state. Basically, devices involving vibration (Staniforth 1982) as well as heap-pouring (Harris and Hildon 1970; Baxter et al. 1997; Graselli and Herrman 1998; Akiyama et al. 1998; Engblom et al. 2012) or shear cells (Tang and Puri 2005) were used to reveal and sometimes model (Tang and Puri 2010) the ability of mixtures to segregate. Most often, these were binary mixtures, widely differing in particle size. This effect has been extensively commented on in the literature (see Vanel et al. 1997) as being the main factor influencing segregation. Particles of different sizes will experience different trajectories during a chute, small particles will be preferably carried by air in fluidized operations or will percolate easily through a coarse particle bed (Rosato et al. 1987). Fewer studies reporting segregation by particle shape have been issued, although they reflect an industrial reality. In the study presented by Massol-Chaudeur et al. (2003), spherical particles demonstrated a clear tendency to segregate as compared to irregular shaped particles of the same size. Spheres are objects of extremely high mobility that can roll over long distances with respect to cubes, however, it is dubious whether they could be chosen as a reliable model to describe particle flow. It is also well known from industrial practice that, if particles are small enough or possess a rough surface, particle-particle interaction may dominate flow and segregation risk may be eliminated. In 2010, Johanson (2010) showed that a mixture of M&M's was subjected to segregation because of a different sugar coating of the yellow "particles" that was actually causing differences in particle surface roughness, producing differences of angle of repose between the particles.

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Reducing segregation potential is, therefore, a matter of reducing differences in the mobility of particles. But mobility is not intrinsic to a particle; it depends on the environment in which it is located, and can be said to be a "mesoscopic" property. Most of the time, segregation results from a collective behavior of the whole set of particles that tends to group together particles of the same nature or exhibiting the same flow properties. This occurs if such particles are particularly different from the rest of the bulk, which is readily the case for binary mixtures. When increasing the number of components of a mixture, even if two components (A and B) are extremely different in mobility, it may happen than A will have similarities with an intermediate particle type (C) that will, in turn, be close to another (D), and so on, up to B. This "chain" globally reduces differences in mobility and provides a sort of mechanical macro-cohesion to the mixture. This is why multicomponent mixtures are empirically known to be less sensitive to segregation than binary mixtures—even if there is only a small difference between the two products. This not only holds for mixtures of particles of different nature (see Tang and Puri 2007), but also for pure products that are constituted of particles of different sizes. It is, therefore, expected that products exhibiting a wide particle size distribution (PSD) range will be less affected by segregation than those having a narrower PSD. In this article, we will confirm this idea by discussing results obtained for two real industrial cases in a heap-pouring test we developed some time ago.

2. Experimental Set up and Methods

2.1 Segregation Device and Powders Used

For this work, we used an altuglass box measuring 3 cm wide, 68 cm long, and 49 cm high (see Massol-Chaudeur et al. 2003 for description). Each powder system was homogenized in a lab-scale double-cone mixer Erweka AR402 for 15 min at a rotational speed equal to 3 rpm (45 revolutions). Powders were then poured from the mixer into the segregation box through a funnel, which formed a heap of small width. The pouring process was controlled by manually adjusting the emptying of the mixer, so as to keep a very small and constant amount of powder in the funnel. Actually, the powder-filling level in the funnel was always limited to the top of the outlet funnel tube, ensuring the mass flow in the funnel and limiting segregation before the formation of the heap. The drop height varied from 40 cm, at the beginning of the experiment, to approximately 10 cm at its end. The device had 48 holes on each side of the box, all the same distance from each other. During heap formation, the holes were plugged so that powder was confined in the test box. Once the heap was formed, plugs were removed and small copper tubes of the same dimensions of the holes were inserted orthogonally to the box, thus, defining samples of an approximate weight of 3 g. All the copper tubes were kept inside the set up during sampling, which was performed by aspiration of the powder in the tubes. Because the set up was transparent, it could be visually assessed that negligible disturbances were caused to the heap during the insertion of the tubes and the sampling process in general. Therefore, we can extract as many samples as holes in the box for further analysis.

Table 1. Some characteristics of the two cases studied. Data were obtained from a Malvern Laser diffraction granulometer for ammonium percholorate and from a Malvern Pharmavision PVS830 for the resin

	d_{10} (μm)	d_{50} (μm)	d_{90} (μm)	$(d_{90}-d_{10})/d_{50}$
Ammonium perchlorate A_1	37.2	215.4	391;9	1.646
Ammonium perchlorate A_2	100.6	215.6	370.9	1.254
Ammonium perchlorate A_3	22.6	196.2	347.9	1.658
Polymeric resin P_1	69.4	160.1	229.5	1.000
Polymeric resin P_2	108.8	190.7	275;71	0.875

Main mechanisms of size segregation involved during the heap formation are as follows (a complete description can be found in Rahman et al. 2011):

- Small particles rolling down the heap are affected by the apparent surface roughness and can be blocked between the spaces left between larger ones before reaching the edge of the heap.
- Coarse particles roll on a smoother surface because their dimension is much higher than that of the fines, so they are less affected by the surface of the heap and can flow down the heap easily.
- Layers consisting of fine or coarse particles are formed alternatively because of the variations of the heap surface caused by the above.

Moreover, rapid flow pattern of particles falling on the top layer of the heap allows the finest particles to percolate between the bigger ones, if the ratio between particles sizes is high enough (1:6.46).

The test was run for two industrial cases, ammonium perchlorate (A) and a polymeric resin (P), each with a different PSD (two cases for P and three cases for A). The physical characteristics on which we base our analysis are presented in Table 1. Sizes in Table 1 are diameters of spheres with equivalent volumes, and PSD are described by volumetric representations. P_1 has a spreader PSD as compared to P_2, which can be diagnosed from the spans of the distributions. Also, it must be noted that P_1 differs from P_2 because it contains many more fine particles (below 100 μm, see Table 2).

Table 2. Volumetric percent of each class of particles (mean over the 23 samples for Polymeric resin, mean over 13 samples for ammonium perchlorate)

Volumetric percent	P1	P2	A1	A2	A3
Class 1 content (<100 μm)	12.29	6.98	10.01	10.53	14.51
Class 2 content	Nd	Nd	50.32	52.71	53.68
Class 3 content	Nd	Nd	31	29.16	26.88
Class 4 content (coarse)	Nd	Nd	8.66	7.59	4.92

A_2 has a narrower PSD than A_1 and A_3. A_3 contains more fine particles and fewer coarse particles than other ammonium perchlorate powders.

2.2 Methodologies

The homogeneity of a mixture is based on the statistical analysis of the content of samples in a key component through a validated sampling protocol. Variance and, more often, coefficient of variation, are classically used to accept or reject a mixture in industrial practice. In the present cases, the aim is to examine if the spread of the PSD of a single product has a positive or a negative influence on the ability to segregate of the powder. This can be studied by analyzing the PSD of the samples taken from the test holes using a criteria based on size classes. The simplest way to do this is to specify a size value x_0 and consider the particles whose sizes are above or below x_0 and that belong to two different species. The content in particles below x_0 in the samples can then be measured and the coefficient of variation calculated. This will be done for the polymeric resin with $x_0 = 100\,\mu m$. To be more precise, we consider various size fractions i as being key components, one after another, and examine each coefficient of variation CV_i. The overall coefficient of variation CV can also be studied if the mean content of each size fractions μ_i is known. This will be done for ammonium percholorate for four size classes (0–100.2 μm; 100.2–251.8 μm; 251.8–399 μm; >399 μm) and the criteria:

$$CV = \mu_1 CV_1 + \mu_2 CV_2 + \mu_3 CV_3 + \mu_4 CV_4.$$

In addition, the degree of segregation calculated hereby must be related to the physics at play. The value of the CV will have a real significance with respect to segregation if the geometrical distribution of the "contents" in the heap demonstrates segregation. In the opposite case, it will be difficult to link the values calculated to a real problem of particle segregation. One also must keep in mind that none of these criteria is absolute, but, if their results all arrive at the same conclusions, it can be considered as evidence that these conclusions hold true.

3. Results and Discussion

3.1 Polymeric Resin Case

For this product, some 23 samples have been defined, taken, and sieved in order to derive their content in fine particles. The two coefficients of variation have been calculated from these data and are given in Table 3. At the scale of the holes (approx. 3 g), both products appear to be segregated with respect to a classic standard of 6%: CV for the distribution of fine content (sizes less than 100 μm) is 9.97% in the case of P_1 and 15.62% for P_2. However, P_1 that has a spreader PSD (span = 1) and contains much more fines than P_2 (see Table 2), also demonstrates a lower CV that means less segregation in the heap. P_2 that has a narrower PSD (span = 0,875) and contains less fines than P_1 (Table 2), also demonstrates a higher CV that means a highest segregation state.

Table 3. Segregation criteria values obtained for each product

CV based on	P1	P2	A1	A2	A3
Class 1 content (<100 μm)	*9.97%*	*15.62%*	6.53%	20.99%	19.57%
Class 2 content	Nd	Nd	1.35%	6.89%	6.36%
Class 3 content	Nd	Nd	1.23%	3.09%	1.5%
Class 4 content (coarse)	Nd	Nd	4.27%	16.30%	19.14
All size fractions			*2.08%*	*7.98%*	*7.60%*

Large particle sizes distributions are known to lead to compact stacks, surely not good for size segregation of particles. This is confirmed by repeated experiments, which are not reported here for clarity reasons.

The visual feeling of the distribution in fine particles in the heaps formed (see Figure 1) confirms this idea. For P_1, there are no poor nor rich zones in fines in the heap, all the samples being close to the mean content. Moreover, the richest samples in fine particles correspond to the edges of the heap. In other words, the distribution of the "compositions" in the tester does not seem to link with the waited segregation phenomena. The opposite situation is obtained with P_2. Segregated zones clearly appear at the edges of the heap (poor fine contents for samples 26, 27, 29, 17, and symmetrically for 38, 37, 37, 25) and at the center of it (rich fine contents for samples 2, 4, 7, 19, 21, 23).

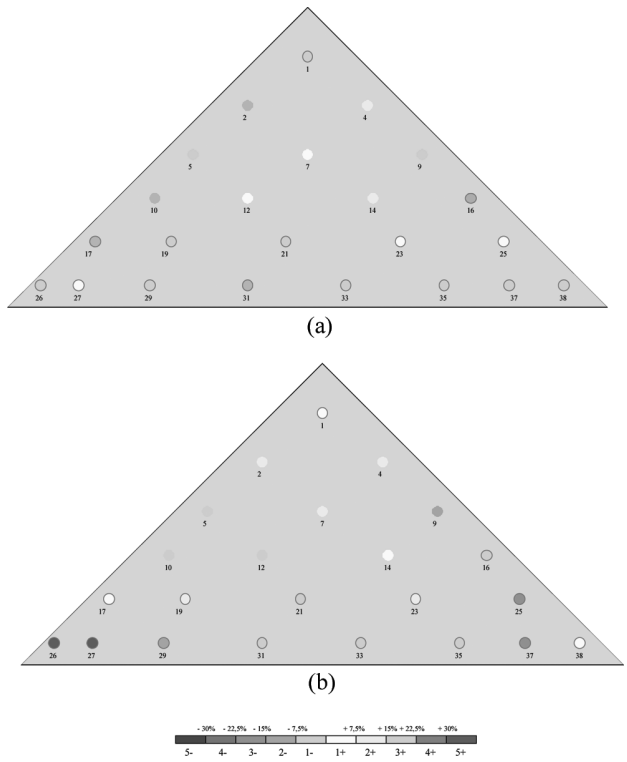

Fig. 1. Repartition of fine particles in the heaps for P_1 (a) and P_2 (b). Colors indicate deviations from the mean content according to the code indicated. (Figure available in color online.)

3.2 Ammonium Perchlorate Case

As previously indicated, the criterion considered for this product is CV based on 4 size fractions. The results are presented in Table 3. Figure 2 illustrates the fine and coarse (size fraction 1 and 4) distribution in the heaps. In Figure 2, one sampling point corresponds to three samples taken in the heap, because the size measurement method (laser diffraction PSD equipment) required 10 g of powder. For A_1, no clearly segregated zones visually appear in the heap, which is confirmed by the CVs calculated. Only a small deviation on fine and coarse contents can be observed.

For A_2 and A_3 the situation is extremely different. The contents in fine and coarse particles in the heap experience a strong deviation from the mean, the related CV being more than four times higher for class 4 and three times higher for class 1. Furthermore, for these size fractions, several individual values of volumetric content are larger than 15% from the mean content value of the heap (see Figure 3). In a usual pharmaceutical standard, these values are unacceptable and the corresponding mixtures should be rejected. A_2 and A_3 look strongly segregated with respect to A_1. As for the previous case, the only difference between the products studied is the particle size distribution.

In each of these three powders, particles between 150 and 250 μm are most represented in the distributions, and small particles with sizes smaller than 40 μm are present in nonnegligible quantity, allowing a percolation pattern to occur. In so far, packing density of the heaps and the pore size distribution in the heaps between particles should play an important role in segregation.

A_2 presents the narrower size distribution (span is only 1.254) and is consequently the most segregation-prone system, which is in accordance with results obtained in polymeric resins cases. Indeed, fine and coarse particles are not well distributed in the heap. The spread of the PSD of A_3 is equal to that of A_1 but this powder contains less coarse particles (only 4.9%; see Table 2.) than other powders. In polydispersed systems, large particles have the higher number of contacts and form contact/power chains in stacks. In this case, it can be argued that the structure of the stacks is due to the largest particles. Therefore, the heap formed by A_3 particles is probably less structured than that corresponding to A_1, allowing more segregation to occur.

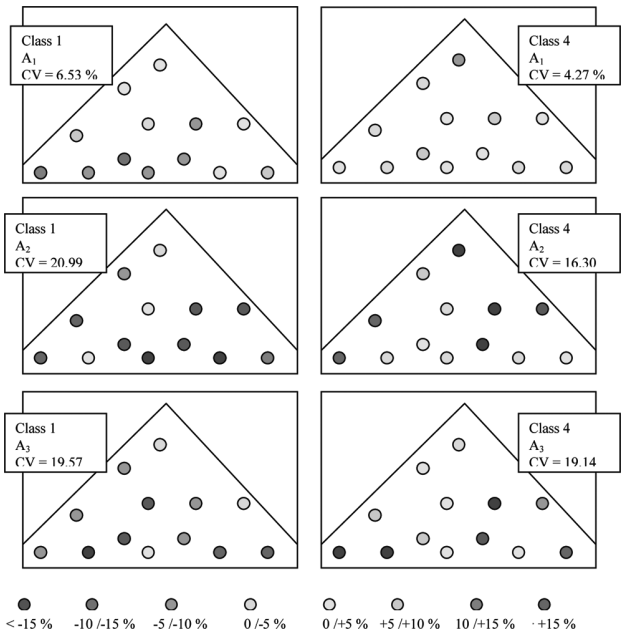

Fig. 2. Distribution of class 1 and class 4 particles for A_1 A_2 and A_3. Colors indicate deviations from the mean content according to the code indicated. (Figure available in color online.)

4. Conclusions

In this study, we used an experimental test to detect different segregation abilities of two industrial products. The results presented demonstrate that segregation can occur due to a particle size distribution effect, and not only due to individual size or shape factors. For the cases studied here, the risk of segregation is real and notable for narrow PSD. It seems that it may be ignored if there is an important spread of the PSD. This has not been noted in the scientific literature so far and goes along with industrial empirical knowledge about multicomponent mixtures. Indeed, the presence of particles of different sizes is relevant to segregation as far as they exist in certain relative quantities. This may be a matter of particle-particle number of contacts, which may reduce mobility and provide this macroscopic apparent cohesion of the particles. A threshold effect may also exist, above which segregation cannot occur, and percolation theory may be adapted to model this very challenging phenomena of granular physics and chemical engineering with industrial application in pharmaceuticals, food products, ceramics, etc. All this will certainly need confirmation, probably using more controllable particulate systems than the real-case systems discussed herein, and certainly through the help of discrete element model (DEM) simulations.

References

Akiyama, A., K. M. Aoki, K. Yamamoto, and T. Yoshikawa. 1998. Experimental study on vibration-induced convection and heaping in granular beds. *Granular Matter* 1:15–20.

Baxter, J., U. Tüzün, D. Heyes, I. Hayati, and P. Fredlung. 1998. Stratification in poured granular heaps. *Nature* 391:136.

Engblom, N., H. Saxen, R. Zevenhoven, H. Nylander, and G. G. Enstad. 2012. Segregation of powder mixtures at filling and complete discharge of silos. *Powder Technology* 215–216:104–116.

Graselli, Y., and H. J. Herrmann. 1998. Experimental study of granular stratification. *Granular Matter* 1:43–47.

Harris, J. F. G., and A. M. Hildon. 1970. Reducing segregation in binary powder mixtures with particular reference to oxygenated washing powders. *Industrial and Engineering Chemistry Process Design and Development* 9:363–367.

Hussain, A. S. 2004. The PAT initiative and the role of pharmaceutical engineering in the 21st century. Presented at the Proceedings of the Albi International Rencontres in Pharmaceutical Engineering, Albi, October, pp. 63–71.

Johanson, K. D. 2010. Understanding angle of repose segregation: characterization of a dual mechanism. Presented at the Proceedings of the AIChE Annual Meeting, Salt Lake City, Utah. Paper No. 477f.

Kim, J. Y., and K. Y. Choi. 2001. Modeling of particle segregation phenomena in a gas phase fluidized bed olefin polymerization reactor. *Chemical Engineering Science* 56:4069–4083.

Massol-Chaudeur, S., H. Berthiaux, and J. A. Dodds. 2003. The development of a static segregation test to evaluate the robustness of various types of powder mixtures. *Transactions of the IChemE C* 81:106–118.

Ottino, J. M., and D. V. Khakhar. 2001. Fundamental research in heaping, mixing and segregation of granular materials: Challenges and perspectives. *Powder Technology* 121:117–122.

Rahman, M., K. Shinohara, H. P. Zhu, A. B. Yu, and P. Zulli. 2011. Size segregation mechanism of binary mixture in forming a conical pile. *Chemical Engineering Science* 66:6089–6098.

Rosato, A., K. J. Strandburg, F. Prinz, and R. H. Swendsen. 1987. Why the Brazil nuts are on top: Size segregation of particulate matter by shaking. *Physical Review Letters* 58:1038–1040.

Staniforth, J. N. 1982. Determination and handling of total mixes in pharmaceutical systems. *Powder Technology* 33:147–159.

Tang, P., and V. M. Puri. 2002. Methods for minimizing segregation: A review. *Particulate Science and Technology* 22:321–337.

Tang, P., and V. M. Puri.. 2005. An innovative device for quantification of percolation and sieving segregation patterns—Single component and multiple size fractions. *Particulate Science and Technology* 23:335–350.

Tang, P., and V. M. Puri. 2007. Segregation quantification of two-component particulate mixtures: Effect of particle size, density, shape, and surface texture. *Particulate Science and Technology* 25:571–588.

Tang, P., and V. M. Puri. 2010. Percolation segregation model for similar and differing constituents. *Particulate Science and Technology* 28:287–297.

Vanel, L., A. D. Rosato, and R. N. Dave. 1997. Size-dependent segregation in a 3D vibrated bed. In *Powders & Grains 97*, eds. R. P. Behringer and J. T. Jenkins. 385–388. Proceedings of the Third International Conference on Powders & Grains, Durham, NC, May 18–23, Rotterdam: A. A. Balkema.

Williams, J. C. 1965. Cause and effects of segregation in powders. *Chemical Processing*.

Williams, J. C. 1976. The segregation of particulate materials—A review. *Powder Technology* 15:245–251.