

## Granular boycott effect: How to mix granulates

J. Duran\* and T. Mazozi

LMDH-UMR 7603 CNRS, ESPCI and University Pierre et Marie Curie, 4 Place Jussieu, 75252 Paris Cedex 05, France

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Granular material can display the basic features of the Boycott effect in sedimentation. A simple experiment shows that granular material falls faster in an inclined tube than in a vertical tube, in analogy with the Boycott effect. As long as the inclination of the tube is above the avalanche threshold, descent of granular material in the tube causes internal convection which in turn results in an efficient mixture of the granular components. By contrast, as in analogous experiments in two dimensions, a vertical fall of granular material occurs via successive block fragmentation, resulting in poor mixing. [S1063-651X(99)02209-6]

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About 80 years ago, Boycott [1] made the surprising discovery that sedimentation of oxalated or defibrinated blood corpuscles occurs five to ten times faster in an inclined tube than in a vertical one. The Boycott effect is now well known and well studied (e.g., [2]). The best explanation of this phenomenon is that sedimentation in a vertical tube requires the supernatant to pass directly through the porous concentrated suspension, which is a slow process. When the tube is inclined, however, the supernatant rises quickly, and easily generates a convective layer in the suspension, as shown in Fig. 1(a).

Our experiments show that dry granular materials display a Boycott-like effect, though based on a different mechanism than in sedimentation. Not only is the fall of granular material faster in an inclined tube than in a vertical one, it also, as in the case of sedimentation, produces an internal convection roll. The convection serves to efficiently mix granular components, which has significant practical consequences.

After water, granulates are the class of material most commonly used in human activity. Thus granulates have been the focus of considerable experimental and theoretical work seeking to describe and understand their static and dynamic properties. From an industrial point of view, some of the central open problems pertain to guided flows in tubes, pipes (e.g., [3]), or hoppers. Also of critical concern is the poorly understood segregation process. In general, mixtures of granular materials tend to separate readily into components differing in size, shape, or micromechanical properties. In many industrial processes, in areas as diverse as pharmaceuticals, ceramics, semiconductors, and polymer handling, this effectively spontaneous segregation is highly undesirable and difficult to avoid. Both our theoretical and experimental work is devoted to the intersection of these open problems, flow in tubes and segregation.

We have presented a theoretical model and computer simulations [4,5] of slowed falling, as well as a possible fragmentation process, in a dense bidimensional granular bed of spherical particles confined between vertical glass walls. This previous theoretical and experimental work points to a unified and detailed description of the process in terms of

*arching*. Arching is the formation of solid contact chains attached to lateral vertical walls; these chains support blocks of granulate material above them. These chains erratically build up and open in the lowest portion of the falling material. The falling velocity decreases due to friction between the granulate and the walls of the tube, but also due to fragmentation processes within the falling material. In a two-dimensional configuration, the friction-limited fall can be summarized as follows: A stack of granular material of height  $h$ , falling through a vertical pipe of width  $D$ , undergoes a reduced downwards acceleration  $\gamma = \Gamma g = (1/\chi)(1 - e^{-\chi})g$ , where  $g$  is the gravitational acceleration, and  $\chi$  is a dimensionless characteristic parameter (known as the decompaction parameter), given by  $\chi = SK\mu$ . Here,  $K$  is the Jans-

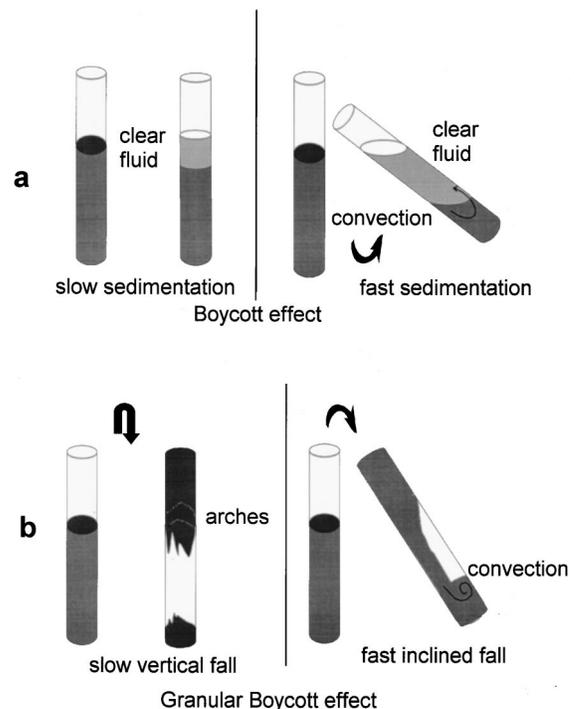


FIG. 1. Sketch showing the analogy between the Boycott effect (a) in sedimentation and the granular Boycott effect (b). In both cases the process is roughly five times faster when the tube is inclined rather than vertical. Convective rolls are present in both cases.

\*Author to whom correspondence should be addressed. Electronic address: jd@ccr.jussieu.fr

sen [6] parameter and  $\mu$  is the Coulomb friction coefficient between the vertical walls and the falling granulate.  $S = 2h/D$  is the shape ratio,  $h$  being the height of the granular column. In many practical situations  $K\mu$  is on the order of 0.2. At this value, a sufficiently tall stack of granular material (e.g., five times as high as wide) confined in a vertical tube is subject to a significantly reduced downward vertical acceleration, with  $\Gamma \ll 1$ . Experiments show that while the material may undergo a progressive fragmentation process, initiated at some wall asperity, this does not contribute to an acceleration of the fall. The process in two-dimensional configuration using monodisperse spherical beads has been the subject of our most detailed analysis to date. However, it appears that experiments in three dimensions using polydisperse granular materials have similar features as reported in [7].

Now, we ask the question, what would occur if, instead of performing the fall along a vertical direction, we were to slant the container (till remaining above the natural avalanche angle which is about  $36^\circ$ ) at an angle lying between  $40^\circ$  and  $60^\circ$  with respect to the horizontal. According to our previous observations, it seems intuitive that because of the inclined walls, the granular material would lean only on one side of the container and thus that the nearly symmetric arch formation which slows down the fall would no longer occur. Thus, we expect a different falling process in the form of fast avalanching.

In a typical experiment, we use a cylindrical leucite tube, 50 cm long and 1 cm in diameter. A large shape ratio  $S$  of the tube is needed to allow sufficient velocity to be reached at the lower end of the tube to produce convective rolls. We fill the tube halfway with glass beads 100–140  $\mu\text{m}$  in diameter. Finer powders seem to give the same results, but potentially due to a different mechanism than that which we wish to study. With a fine powder, the fall may be slowed due to atmospheric drag, and thus by a mechanism quite similar to that of the original Boycott effect in solid-liquid sedimentation.

The experimental sequence is sketched in Fig. 1(b). (1) The tube is held vertically and filled halfway with granular material. (2) The tube is quickly inverted, and while being held vertically, the material is allowed to fall out. As we have previously reported [7], the material does not fall uniformly, but rather, the upper part of the packed material remains relatively stationary, while the lower part falls first. In addition, fall from within a tube is slower than free fall. The tube empties within 1 to 3 s, depending on the material used,

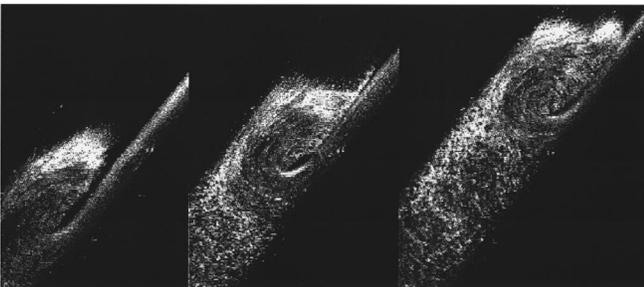


FIG. 2. A sequence of snapshots at 20-ms intervals of the convective roll in an inclined tube of granulate. The flow is from right to left.

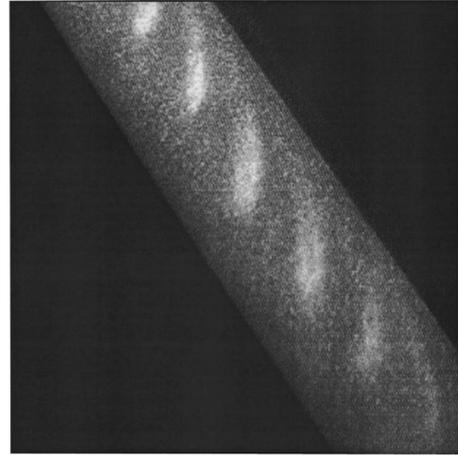


FIG. 3. A luminescent trail left in an inclined tube after a Boycott effect experiment. At the beginning of the experiment, the luminescent powder lies on top of the granular material. This long-time-exposure image was taken after a single rotation of the cylinder. Illumination in the UV shows white zones indicating a high concentration of luminescent powder. Tracking the powder in this way, we find that it is progressively mixed into the granular material.

and the degree to which the preparation is compacted [7]. To compare the vertical fall behavior with fall from an inclined tube, we begin as above with a vertical, half-filled tube, and quickly rotate it to a predefined angle of  $45^\circ$ – $60^\circ$  with respect to the horizontal. At these angles, the tube empties within 300 ms. The granulate falls in a continuous avalanche. Note that the relationship between the vertical fall duration and the inclined fall duration is scale dependent. The vertical

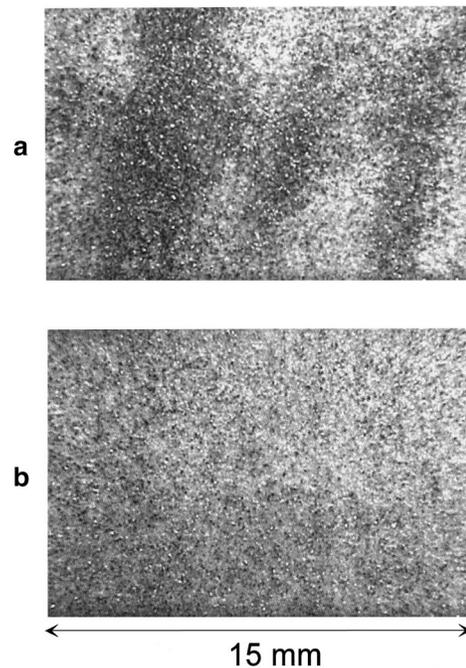


FIG. 4. Illustration of efficient mixing properties of the ascending roll after five rotations of a mixture of different sizes and dark (large) and white (small) particles. Snapshot (a) was taken at the beginning and (b) at the end of the process. A careful analysis of the final product shows that the mixing is practically perfect.

fall duration scales as  $(h/\Gamma)^{1/2}$  and thus can be significantly larger than the inclined fall duration, which scales as  $h^{1/2}$ . A careful examination of the process using a charge coupled device (CCD) camera (Fig. 2) shows the existence of a strong convective roll at the interface between the avalanche stream and the previously deposited granulate. To better view this process, we use a granular material made of submillimetric glass beads and submillimetric luminescent Mn-doped ZnS particles. The luminescent powder is deposited on top of the glass powder. We perform an experiment as above in the dark, and after one rotation cycle we illuminate the tube in the ultraviolet. The luminescent powder displays a roughly cycloidal trajectory as seen in Fig. 3. Through the course of successive experiments using the same mixture repeatedly, the granulate is rapidly well mixed. The successive cycloidal trajectories do not coincide, producing mixture throughout the material. This mixing property may have great industrial impact. Compositions of particulate solids must often be well mixed before they can be further treated in an industrial process, such as fusion, or a chemical reaction [8]. At present, mixing is typically accomplished by introducing a stirring device, such as an Archimedes screw, into the composition. The device mixes by generating local

convection, and thus must be moved throughout the material to produce complete mixing. By contrast, using the granular Boycott effect one can mix material in a sealed container. This may be the key feature in any process involving dangerous, reactive, or sensitive chemicals. In a practical context, it is important to note that the granular Boycott effect leaves a thin layer of the granulates in contact with the tube during the fall unmixed, since this layer is not reached by the convective roll. This problem can be addressed by applying the procedure successively as required, rotating the tube around its long axis between applications. In this way, we were able to obtain industrial-grade mixing of large black and fine white granular powders (Fig. 4). We noted earlier that mixing by the granular Boycott effect is scale dependent. To show that the procedure is also effective on an industrial scale, we performed a series of experiments as above, but using a 2-m tube. In this way we obtained good mixing of various cereals and chemical powders in sealed tubes. Still larger scale experimentation is underway.

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