Linking compaction dynamics to the flow properties of powders

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We have investigated the flow properties of powders by using two classical techniques based on the shear stress measurements and the count of intermittent avalanches respectively. Results are compared with measurements of the compaction dynamics. Strong correlations are evidenced between compaction relaxation parameters and free flow characteristics. Those correlations are given by semi-empirical laws based on physical arguments. This work opens perspectives in powder technology and the knowledge on granular matter.

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The complex collective behaviors of an assembly of particles is due to the dissipative nature of the inter-particle forces. A wide variety of phenomena [1] can be observed by varying the strength of power supply to a granular system. A low power input may induce slow relaxation in a series of metastable states (granular compaction [2, 3]) or an intermittent collective motion (stick-slip [4–6]), while if the energy supply is large enough a steady state (free flow [7]) will result. Those dynamical regimes are illustrated in Figure 1. The physical properties of powders are mainly due to the interplay of cohesive forces and inter-grain frictional forces. Indeed, the formation of large grain aggregates and the presence of arches in the system prevent a powder to flow. Poor flowability of bulk material causes a number of problems in the production of solid dosage forms. The measurement of flowability in the pharmaceutical field has been shown to be very critical in the past [8]. The knowledge of powder properties has a major economical impact on all industries.

In the present letter, we propose to set a bridge between an original measurement of compaction dynamics and the flow properties of powders. Both aspects are often treated separately. The three dynamical regimes cited above have been experimentally investigated with different methods. Different powder mixtures containing Riboflavine (vitamin), Avicel® (micro-crystalline cellulose used as excipient) and Aerosil200® (colloidal silicon dioxide) have been considered in order to test a broad range of behaviors from poor to high flowability. Those powder mixtures are commonly encountered in pharmaceutical industries. *Slow compaction* – The Carr's index [8, 9] based on compaction is commonly used since many years to determine the flowability of powders. However, this test is often criticized for his lack of reproducibility and accuracy. Indeed, this test is based exclusively on two volume fraction measurements made with naked eye : the initial volume fraction and the volume fraction after 500 taps. In this paper, we propose to use an experimental apparatus developed earlier to perform fundamental studies on granular materials [10, 11]. We follow the height of a column of granular materials when an electromechanics hammer produces a series of identical taps at the bottom of the system. The height is then converted into volume fraction η which is monitored as a function of the tap number n. The relevant parameter extracted from compaction curves is the relaxation time τ of that process by fitting compaction curves. One has

$$\eta = \eta_0 + \frac{(\eta_\infty - \eta_0)}{1 + \ln(1 + \frac{n}{\tau})} \tag{1}$$

This logarithmic compaction law has been proposed in an earlier work [2]. It could be obtained within a kinetic theory which includes a "caging effect" [3] for grains. For powders, the logarithmic law fits better the data than the Avrami (exponential) law [11, 12], proposed for non-cohesive granular materials. The latter has been recently demonstrated within a kinetic theory [13] in the high packing fraction limit, never reached by powders. *Shearing* – In the classical shearing measurements (as described by Jenike in [14]) the flowability is determined in an annular cell. This method is well established in chemical engineering, in particular for the design of silos and blenders. The F value (flowability index) is defined as the ratio of the consolidation stress σ_1 and the unconfined yield strength f_c. One has

$$F = \frac{\sigma_1}{f_c} \tag{2}$$

Depending on the F index value, the powder could be classified in a scale of flowability [15] : $(F < 1)$ no flow, $(1 < F < 2)$ highly cohesive powder, $(2 < F < 4)$ cohesive powder, $(4 < F < 10)$ intermittent flow, $(F > 10)$ free flow. *Avalanches* – Another measurement described in the European Pharmacopoeia [8] is the count of avalanches in a rotating drum when the angular velocity is fixed. The relevant parameter is the number of avalanches N per rotation when the container motion is slow. Herein, we fixed the angular velocity to $\omega = 0.046$ rad/s. Thus, we have three different techniques for probing a large set of pharmaceutical powders. Those three techniques involve different scales for grains displacements in the materials : small grain displacements less than a grain diameter for each stage of compaction, grain displacements being the order of arches (shearing), high grain displacements during free flow.

Results – Figure 2 exhibits a typical compaction curve of a powder fitted with Eq.(1). The agreement is good. Please note the low packing fraction $0.20 < \eta < 0.40$ indicating that cohesive forces dominate gravity. Those values of η are quite different from granular non-cohesive systems for which $0.45 < \eta < 0.7$ [10]. The relaxation parameter τ multiplied by $e - 1$ is shown on the plot and represents the number of taps to reach the packing fraction $\eta = (\eta_0 + \eta_\infty)/2$.

Figure 3 presents the relaxation time τ as a function of the flowability parameter F. A clear correlation is observed within error bars. A high flowability involves a small relaxation time, i.e. a fast compaction system. On the contrary, a granular system exhibiting a slow compaction dynamics has poor flow properties. If we assume that the characteristic force needed to initiate a grain motion is close to the yield stress f_c, the time needed to obtain a free displacement of one grain diameter

d is proportional to $\sqrt{d/F}$ from the classical kinematics. A cutoff at $F = 1$ should be considered in order to define a limit for flowing. Superposing this characteristic time to the natural relaxation time τ_0, one has

$$\tau = \tau_0 + \frac{B}{\sqrt{F-1}} \tag{3}$$

This semi-empirical law is shown fitting the data in Figure 3. The parameter B contains various grain characteristics such as particle size, density, etc. We obtained $\tau_0 = 11.6 \pm 1.9$ and $B = 31.7 \pm 6.2$ for both free fitting parameters. There is a strong link between shearing and compaction. Of course, a more detailed model should be developed but this is left for future work. The dispersion of collected data around the trend of Eq.(3) suggests that one parameter is not enough to describe precisely the flow properties. Indeed, it is known that the volume fraction represents only a part of the relevant parameters for describing memory effects [16] and other complex phenomena [13] discovered recently in granular materials.

Figure 4 presents the number of avalanches N per drum rotation as a function of the relaxation time τ for compaction. The number of avalanches is of course quite low when the powder is cohesive. The number of avalanches increases when the powder compacts faster. For cohesive granular systems like the powders we investigated, the avalanches are mainly clusters of grains rolling on the surface rather than grain layers. The fragmentation of the powder into clusters has a characteristic time related to the one for compaction. We propose that the avalanche rate scales as

$$N = \frac{b}{\tau} \tag{4}$$

which is fitted in Figure 4 on the data. The agreement is good. From that fit, we obtain $b = 623 \pm 15$. This value could be interpreted as the upper boundary value for τ in the case of extreme cohesive system. The relationship (4) evidences a clear link between avalanches and compaction. Indeed, shearing and compaction are treated as separate phenomena.

One should remark that the classical measurement of the angle of repose θ seems to be independent from the three above properties (τ, F and N). Contrary to common belief, this zero correlation is not so surprising since this concerns a comparison between static and dynamical properties of packings. Most of the established methods (e.g., angle of repose, critical orifice diameter) [8] do not directly reflect flowability but summarize a mixture of different physical parameters, and do not take into account relaxation mechanisms of the material in the container. Since a strong correlation exists between flow of powders and compaction, it means that physical parameters are responsible of granular properties. Those parameters should have a microscopic origin since they have already an effect for very small grain displacements. Grain mobilities [11] should play an important role.

The great advantage of compaction measurements over flow measurements are the high reproducibility of collected data and the easy preparation of samples. From a fundamental point of view, the correlation between extremely slow relaxation and flows is important for testing models of granular systems, even for non-cohesive ones. In summary, we have evidenced strong correlations between compaction and flow properties of powders. Semi-empirical laws have been proposed.

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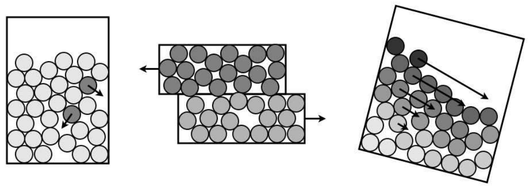

FIG. 1: Sketch of three dynamical regimes for granular systems. Colored grains are moving objects. Arrows indicate grain motions. (left) When submitted to series of taps, a packing experiences a slow compaction due to small displacements of grains. (center) The shearing of granular systems induces an intermittent breaking of arches. (right) The free flow of a granular system, when enough power input is injected in the system, involves a dynamics of avalanches and strong velocity gradients in layered motion.

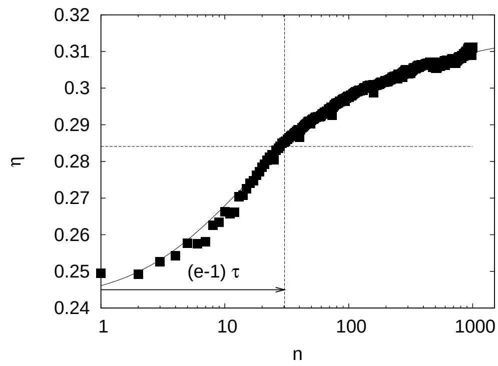

FIG. 2: Typical compaction curve of a powder in a semi-log plot. The continuous curve is a fit using Eq.(1). The relaxation parameter τ multiplied by $e-1$ is shown on the plot and represents the number of taps to reach the packing fraction $\eta = (\eta_0 + \eta_\infty)/2$.

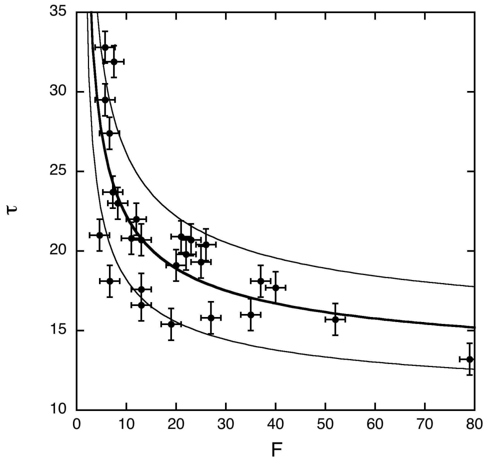

FIG. 3: The relaxation parameter τ as a function of the flowability index F for various powders. Error bars on measurements are indicated. The thick curve is a fit using Eq.(3). Both thin curves represent the limits of the interval of confidence of Eq.(3).

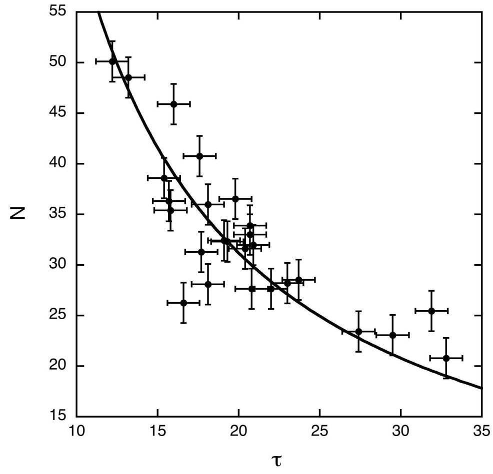

FIG. 4: The number N of avalanches in a rotating drum as a function of τ for various powders. Error bars on measurements are indicated. The curve is a fit using Eq.(4).