UDK: 624.131.52.001.3:544.6:335
COSATI: 11-07

Compaction Dynamics of Vibrated Granular Materials

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The vibratory compaction of granular materials has long been of importance in technological applications. A number of diferent approaches have been proposed in order to connect slow compaction with effects of excluded volume and geometrical frustration without reaching a unique conclusion on the temporal behaviour of density change. When analyzing the previous experimental results, we found that the density relaxation of a vibrated granular material follows the Mittag-Leffler law in the whole temporal range. Furthermore, a fractional model of granular compaction that captures this relaxation dynamics is presented. This paper provides some of the relevant background and attempts to identify some of the research needs from an industrial perspective.

Key words: **granular materials, granular compaction, fraction kinetics, material properties, experimental results, military application.**

Introduction

MANY fields share an interest in granular materials [1]. There is a great need to know if the snow on a mountain slope is stable, or how grain will flow in a silo. Practically everything we eat started out in a granular form. Granular materials play an important role in industries such as mining, agriculture and construction. They are also important in geological processes such as landslides and erosion and, on a larger scale, plate tectonics which determine much of Earth's morphology.

Granular materials are simple. They are large conglomerations of discrete macroscopic particles. If they are noncohesive, the forces between them are strictly repulsive. The particles are usually surrounded by a fluid, most often air, which may play a role in the dynamics of the systems. Examples of such materials include sand, stones, soil, ores, grains, pharmaceuticals, and a variety of chemicals.

At the root of the unique status of granular materials are two characteristics: ordinary temperature plays no role, and interactions between grains are dissipative because of the existence of static friction and the inelasticity of collisions. There are no long-range interactions between individual grains or between individual grains and the walls of a confining container. Granular matter refers to particle systems in which the size is larger than one micron. Below one micron, thermal agitation is important, and Brownian motion can be seen. Above one micron, thermal agitation is negligible. We are interested here in many-particle systems, at zero temperature, occupying a large variety of metastable states: if we pour sand on a table, it would tend to go to a ground state, with a monolayer of grains giving the lowest gravitational energy. However, in reality, the sand remains as a heap; the shape of the heap and the stress distribution inside depend critically on how the heap was made, i.e., a static pile will remember how it was formed [2]. Thus, any particular grain configuration in the material corresponds to a metastable state that will persist indefinitely until it is disturbed by some (nonthermal) means.

And yet, despite this seeming simplicity, a granular material behaves differently from any other familiar form of matter – solids, liquids, or gases – and should therefore be considered as an additional state of matter in its own right. Let us consider these differences.

A common gas: Highly agitated systems of particles are often modeled in a manner similar to a rarefied gas [3]. An important difference is that, unlike collisions between gas molecules, solid particle collisions are inelastic and dissipate energy, so that a "gas" of particles will slow down and come to rest in clumps. As a result, novel features arise for the statistical mechanics of these systems. Any seemingly fluidlike behaviour of a granular material is a purely dynamic phenomenon. For example, the surface waves do not arise as a linear response to external energy input but are the consequence of a highly nonlinear hysteretic transition out of solidlike state.

A common fluid: Granular materials also have liquid-like properties. For example, if a sand pile is tilted several degrees above the angle of repose, grains start to flow. However, this flow is clearly not that of an ordinary fluid because it only exists in a boundary layer at the pile's surface with no movement in the bulk at all.

A common solid: For example, when granular material is held in a silo, the pressure head is not height-dependent as it would be in a normal fluid; that is, the pressure at the base of the container does not increase indefinitely as the height of the material inside it increases. Instead, for a sufficiently tall column, the pressure reaches a maximum

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value independent of the height. Because of the contact forces between grains and static friction with the sides of the container, the container walls support extra weight.

Motivation for studying granular materials

If we measure it by tons, the material most manipulated by man is water; the second most manipulated material is granular matter. Particle technology spans a range of product lines:

- "clinkers" (the starting point of cement) are complex mixtures of silicoaluminates, calcium silicates, etc.
- "builders" are an important part of commercial detergents: they are based on inorganic particles such as calcium carbonate.
- most pharmaceutical products are derived from powders, obtained by precipitation, crystallization, or prilling.

In the chemical industry alone, 60% of products are manufactured as particulates and further 20% use powders as ingredients to impart specific end-use properties [4]. The US production presently accounts for over a trillion kilograms each year of granular pharmaceuticals, foods, and bulk chemicals [5]. Despite its importance, however, our understanding of granular processes is limited and manipulations of powders still involve some very clumsy and/or dangerous operations:

- Milling is slow, inefficient, and generates a very broad distribution of final sizes.
- The smaller-size component of these distributions is often toxic.
- Many powders, when dispersed in the air, achieve a composition that is ideal for strong detonations.

The manipulation of mixtures is delicate. Numerous mechanisms for segregation [6] of dissimilar grains have been cataloged, including percolation, convection, inertia, ordered settling, and arching. Currently, there is no general theory that shows how these many mechanisms are related or under which conditions one or another mechanism will dominate. Even the way in which powders are loaded into blenders of common design can alter the time needed to homogenize them by as much as two orders of magnitude. As a result, factories that rely on powder handling require much longer start-up times than typical plants involving only fluids.

At present, physicists do not know how to predict a priori whether two powders will mix or segregate when stirred together in a given blender. Convection is far faster and more efficient mixing mechanism in grains (as in fluids). Diffusion is much slower than convection, but it occurs in all directions. Thus, in the absence of segregational tendencies between dissimilar particles, diffusion will eventually lead to a completely homogeneous mixture. When diffusion dominates, the mixing problem is reduced to that of finding the parameters that minimize the blending time. Better understanding of the transport process should help in predicting whether a given flow will mix or segregate its constituents.

The dynamics of penetration in granular materials is important to a variety of military applications. Some researchers seek to develop better ways of holding deeply buried targets at risk. It is not clear how mechanisms of penetration in granular materials depend on factors such as the impact velocity, packing of the material, particle size distribution and other material properties. Predictive models of the particle dynamics in such problems are limited because the fundamental mechanisms of the interaction between a penetrator and granular media, as well as the dynamics of granular matter, are not well understood. Modeling granular materials could also help protect military vehicles. Transparent ceramic armor - heavy, see-through material mounted as a protective windshield on tanks and other vehicles behaves like a granular material when hit by a projectile such as a bullet, radiating pressure away from the damaged area.

We see the importance of fundamental research in granular matter. This was appreciated very early in mechanical and chemical engineering; physicists have joined in more recently. For them, granular matter is a new type of condensed matter, as fundamental as a liquid or a solid. The aim of engineers is to reproduce phenomena or to control phenomena, while our aim is to construct a statistical or fluid mechanics of the cooperative dynamics of powder. We also aim to determine the mathematical structure behind various kinds of powder behavior. Although these aims are not directly related to engineering questions, we hope they can be the first step toward understanding the fundamental properties of granular matter which is needed by engineers.

In the next section, we present and discuss the results regarding temporal evolution of the packing fraction $\rho(t)$ during the compaction process. It is found that the density relaxation of a vibrated granular material follows the Mittag-Leffler law in the whole temporal range. Furthermore, a fractional model of granular compaction that captures this relaxation dynamics is presented.

Granular compaction

The phenomenon of granular compaction involves the increase of the density of granular material subjected to shaking, tapping or, more generally, to some kind of external excitation. Various experimental studies have underlined the fact that the dynamics of granular compaction is a complex problem. The first experiments [7] have shown that the density compaction under tapping follows an inverse logarithmic law with the tapping number, $\rho(\infty)-\rho(t)\sim\frac{1}{\ln(t)}$. More recently, Bideau and co-workers [8, 9] showed that the compaction dynamics is consistent with the stretched exponential law:

$$\rho(t)=\rho_\infty-(\rho_\infty-\rho_0)\exp\left[-\left(\frac{t}{\tau}\right)^\beta\right]. \quad (1)$$

Here ρ_0 is the initial packing fraction and ρ_∞ is the mean value of the packing fraction at the stationary state.

A number of different models have been proposed in order to identify the physical principles underlying granular compaction [10–13] without reaching a unique conclusion concerning the temporal behavior of the density change. However, when analyzing the previous experimental results [7–9], we found that the Mittag-Leffler behavior (Eq. (2)) describes excellently the compaction dynamics in the whole temporal range. Our motivation arises from the fact that the Mittag–Leffler function is one of the most frequently used phenomenological fitting functions for non-Debye relaxation processes in many complex disordered systems such as metallic glasses, spin glass alloys, ferroelectric crystals and dielectrics [14].

The fitting function that we have used is:

$$\rho(t) = \rho_\infty - \Delta\rho E_\beta\left[-\left(\frac{t}{\tau}\right)^\beta\right], \quad \Delta\rho = \rho_\infty - \rho_0, \qquad (2)$$

where ρ_∞, ρ_0, τ and β are the fitting parameters. E_β denotes the Mittag-Leffler function of the β order. It is defined through the inverse Laplace transform:

$$E_\beta\left[-\left(\frac{t}{\tau}\right)^\beta\right] = L^{-1}\left\{\left(u + \tau^{-\beta}u^{1-\beta}\right)^{-1}\right\}, \qquad (3)$$

from which the series expansion:

$$E_\beta\left[-\left(\frac{t}{\tau}\right)^\beta\right] = \sum_{n=0}^{\infty}\frac{\left(-\left(\frac{t}{\tau}\right)^\beta\right)^n}{\Gamma(1+\beta n)}, \qquad (4)$$

can be deduced; in particular, $E_1\left(-\frac{t}{\tau}\right) = \exp\left(-\frac{t}{\tau}\right)$. The Mittag–Leffler function interpolates between the initial stretched exponential form:

$$E_\beta\left[-\left(\frac{t}{\tau}\right)^\beta\right] \sim \exp\left[-\frac{1}{\Gamma(1+\beta)}\left(\frac{t}{\tau}\right)^\beta\right], \quad t << \tau, \qquad (5)$$

and the long-time power-law behavior:

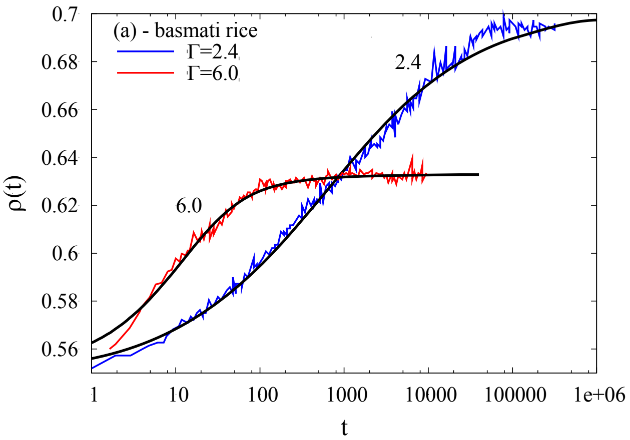
$$E_\beta\left[-\left(\frac{t}{\tau}\right)^\beta\right] \sim \frac{1}{\Gamma(1-\beta)}\left(\frac{t}{\tau}\right)^\beta, \quad t >> \tau. \qquad (6)$$

From Eqs. (5), (6) and (2), one obtains:

$$\rho(t) \sim \rho_\infty - \Delta\rho \exp\left[-\frac{1}{\Gamma(1+\beta)}\left(\frac{t}{\tau}\right)^\beta\right], \quad t << \tau \qquad (7)$$

$$\rho(t) \sim \rho_\infty - \Delta\rho\frac{1}{\Gamma(1-\beta)}\left(\frac{t}{\tau}\right)^{-\beta}, \quad t >> \tau \qquad (8)$$

In Ref. [9], the authors analyzed the effect of the grain anisotropy on granular compaction under vertical tapping and reduced lateral confinement. They observed that the main relaxation features of granular compaction do not qualitatively depend on the grain shape. We produced a good fit of data from Ref. [9]. Our fits are shown in Fig.1 for two tapping intensities Γ, and the fitting parameters are reported in Table 1.



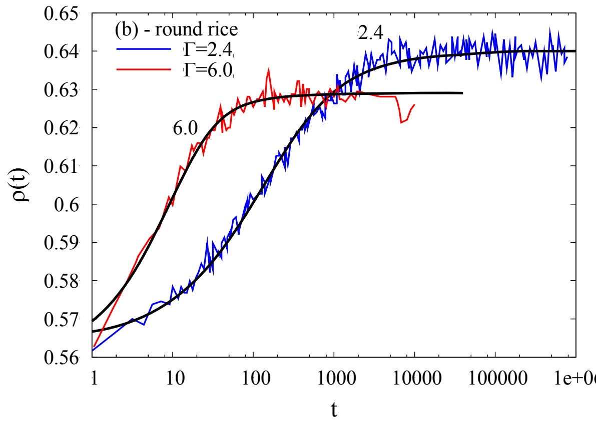


Figure 1. Experimental data from Ribière et al. [9] on the temporal evolution of the mean volume fraction $\rho(t)$ of (a) basmati rice (long grains) and (b) round rice (short grains) for different tapping intensities, Γ =2.4 and 6.0. The continuous superimposed lines are fits according to Eq.(2). The parameters of the fit are reported inTable I.

Table 1. Fitting parameters from Eq. (2) for the experimental data of Ribière et al. [9] on the volume fraction relaxation $\rho(t)$ presented in Fig.1. The anisotropic grains used are of two different shapes: long grains(basmati rice) and short grains(round rice)

	Γ	ρ_∞	ρ_0	τ	β
(basmati rice)	2.4	0.702	0.548	1004.0	0.440
(round rice)	2.4	0.640	0.564	181.0	0.665
(basmati rice)	6.0	0.633	0.552	15.0	0.750
(round rice)	6.0	0.629	0.560	10.5	0.838

Recently, we carried out the extensive simulations of compaction dynamics of frictional hard disks in two dimensions, subjected to vertical shaking [15]. Shaking is modeled by a series of vertical expansions of the disk packing, followed by dynamical recompression of the assembly under the action of gravity. The second phase of the shake cycle is based on an efficient event-driven molecular-dynamics algorithm. We analyzed the compaction dynamics for various values of the friction coefficient and the coefficient of normal restitution. We found that the time evolution of the density is described by the Mittag-Leffler function (2). The parameter τ is found to decay with the tapping intensity Γ according to the power law $\tau \propto \Gamma^{-\gamma}$, where the parameter γ is almost independent on the material properties of grains.

Our simulation results for the deposition of extended objects on a triangular lattice [16] might indicate that the compaction law (2) is universal in the sense that it holds for any shape of granular objects. Therefore, it needs to be clarified whether the Mittag-Leffler law is just a good fit to the experimental data or it has a deeper meaning.

Fractional model of granular compaction

It is interesting to note that the fitting function (2) is a solution of the fractional kinetic equation [17]:

$$\frac{d}{dt}\Delta\rho(t) = -\tau^{-\beta}\,{}_0D_t^{1-\beta}\Delta\rho(t), \quad 0 < \beta < 1, \qquad (9)$$

where $\Delta\rho(t) = \rho_\infty - \rho(t)$. The operator ${}_0D_t^{1-\beta}$ is the Riemann–Liouville (R-L) operator of fractional integration:

$$ {}_0D_t^{1-\beta}\Delta\rho(t)=\frac{1}{\Gamma(\beta-1)}\int_0^t (t-t')^{\beta-2}\Delta\rho(t')dt' . \quad (10)$$

The R-L operator introduces a convolution integral into Eq.(9) with the power-law kernel $M(t) \propto t^{\beta-2}$. Therefore, the fractional kinetic equation (9) involves a slowly decaying memory, so the present density $\rho(t)$ of the system depends strongly on its history $\rho(t')$, $t'<t$. This is in accordance with the fact that granular materials are intrinsically non-local.

In order to obtain a further insight into possible applications of fractional relaxation equation (9) to granular compaction, we imagine an artifical, but instructive model of a powder similar to the 'two-volume' model proposed by Edwards [18]. Our model aims to imitate, in a very simplified way, the compaction of granular material under weak tapping. We suppose that there are only two possible configurations of grains. Grain in the state ↓ (↑) is 'well oriented' ("not well oriented"), which means that a surrounding void space is minimal (maximal). Let N be the total number of grains, $N_\downarrow(t)$ the number of grains in the state ↓ and $N_\uparrow(t)$ the number of grains in the state ↑, so that $N=N_\downarrow(t)+N_\uparrow(t)$. Let us denote by $p^\downarrow(t)$ and $p^\uparrow(t)$ the probabilities to find the grain in the states ↓ and ↑, respectively. Thus we write the packing fraction $\rho(t)$ as:

$$\rho(t)=\rho_\downarrow p^\downarrow(t)=\rho_\uparrow p^\uparrow(t)=\rho_\uparrow+(\rho_\downarrow-\rho_\uparrow)p^\downarrow(t) . (11)$$

We have two limits: $\rho=\rho_\downarrow$ when $p^\downarrow=1$ (free volume is minimal) and $\rho=\rho_\uparrow$ when $p^\downarrow=0$ (free volume is maximal). Suppose now that the master equation $\frac{dp}{dt}=\hat{\omega}p(t),\ p\equiv\left[p^\downarrow p^\uparrow\right]^T$ models the dynamical behavior of the granular system submitted to vertical vibration. The transition rate operator $\hat{\omega}$ would be a function of the strength of the vibration, Γ. The master equation can be written in the integral form:

$$p(t)=p(0)+\int_0^t d\tau\,\hat{\omega}\,p(\tau) . \quad (12)$$

During the tapping, the grains in the bulk experience the external perturbation as a random force. The grains have some freedom to rearrange their positions relative to their neighbors. If the intensity of vibration is sufficiently small, some grains are not able to break away from their clusters, so structures such as bridges are long-standing even during tapping. A major mechanism of compaction is the gradual collapse of these long-lived bridges [19], resulting in the disappearance of the void space which is trapped in the arches ("bridge collapse"). Therefore, the change of a certain configuration occurs due to cooperative rearrangement of free volume between the neighboring grains. The main physical idea of our approach is that the time intervals between the consecutive grain's jumps ↓→↑ (or ↑→↓) are governed by a waiting-time distribution ψ(t). The ψ(t) distributions may stem from possible obstacles and traps that delay the grain's jumps and thus introduce the memory effects into the compaction. If the mean waiting time $T=\int t\psi(t)dt$ diverges, as is the case for the power-law waiting-time distributions of the form $\psi(t)\propto\left(1+\frac{t}{\tau}\right)^{-(1+\beta)}$, $0<\beta<1$, then Eq. (12) generalizes to the fractional form [20]:

$$\begin{bmatrix} p_\beta^\downarrow(t)-p^\downarrow(0) \\ p_\beta^\uparrow(t)-p^\uparrow(0)\end{bmatrix}=\begin{bmatrix} +\frac{\omega}{2} \\ -\frac{\omega}{2}\end{bmatrix}\frac{1}{\Gamma(\beta)}\int_0^t d\tau\frac{p_\beta^\uparrow(\tau)-p_\beta^\downarrow(\tau)}{(t-\tau)^{-(\beta-1)}} . (13)$$

Here, the transition rates $\omega/2$ and the constant β do not depend on the time t, but can depend –at least in general – on the vibration intensity Γ of the shaking process and on the structural properties of a granular system such as grain size, grain material, grain shape, etc. Therefore, a deeper investigation into this point is needed and we hope to stimulate some future experimental and theoretical works. Finally, the fractional evolution equation (9) with $\tau=\omega^{-1/\beta}$ follows from Eqs. (11) and (13) and the steady state condition $p_\beta^\downarrow(\infty)=p_\beta^\uparrow(\infty)=\frac{1}{2}$. It should be noted that the non-Markovian time evolution of the packing fraction is a direct consequence of the random time steps belonging to the long-tailed waiting-time distribution $\psi(t)\sim t^{-1-\beta}$, $0<\beta<1$. Granular compaction is an example of a non-local temporal phenomena in which a different kind of calculus, i.e. fractional calculus, should play a central role.

Concluding remarks

It is found that the density relaxation of a vibrated granular material follows the Mittag-Leffler law in the whole temporal range. Furthermore, a fractional model of granular compaction that captures this relaxation dynamics is presented. The simplicity of our model makes it a useful conceptual tool for probing the dynamical responses of vibrated granular material. Our model is exactly solvable, which allows one to describe the by now well-established picture of Mittag-Leffler compaction, in terms of two parameters, τ and β.

While the unprecedented growth of computer power provides a realistic possibility that simulation techniques will allow the efficient selection and optimization of products and processes, significant advances in experimental, theoretical and computational procedures are required. In particular, this will require an interdisciplinary approach in order to understand and characterize the wide spectrum of behavior exhibited at the continuum and small scales by particulate materials. Industry requires commercial software codes that are well documented and supported. Industry also requires measurement and data interpretation procedures for obtaining the physical, thermal and mechanical information required as an input to the codes. There is a need for considerable research on procedures for obtaining consistent bulk and wall constitutive relationships and the associated material parameters.

Acknowledgments

This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia, under Grants No. ON171017 and III45016 (2011.-2014.).

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Received: 15.08.2012.

Dinamika kompaktifikacije vibrirajućeg granularnog materijala

Vibraciona kompaktifikacija granularnih materijala je od velike važnosti u tehnološkim primenama. Korišćeni su različiti pristupi da bi se u vezu dovela spora kompaktifikacija sa raznim geometrijskim efektima, bez uspostavljanja jedinstvenog zaključka o prirodi vremenske evolucije gustine materijala. Analizom prethodnih eksperimentalnih rezultata, našli smo da relaksacija gustine granularnog materijala sledi Mittag-Leffler-ov zakon u celom vremenskom opsegu. Osim toga, prikazan je frakcioni model granularne kompaktifikacije koji je u skladu sa ovim zakonom. Ovaj rad pokušava da identifikuje pravce istraživanja svojstava granularnih materijala na osnovu potreba industrije.

Ključne reči: **granularni materijali, granularna kompaktifikacija, frakciona kinetika, osobine materijala, eksprimentalni rezultati, vojna primena.**

Динамика компактификации вибрирующих гранулированных материалов

Вибрирующая компактификация гранулированных материалов имеет большое значение в технологических приложениях. Различные подходы были использованы для достижения связи медленной компактификации с различными геометрическими эффектами, без создания общего вывода о природе времени эволюции плотности материала. При помощи анализа предыдущих результатов экспериментов, мы обнаружили, что релаксация плотности гранулированного материала следует закону Миттаг-Леффлера во всём диапазоне времени. Кроме того, ещё показана дробная модель гранулированной компактификации, которая в соответствии с настоящим законом. Эта статья пытается определить направления исследований свойств гранулированный материалов на основе потребностей промышленности.

Ключевые слова: **гранулированный материалы, гранулированная компактификация, дробная кинетика, военные применения.**

La dynamique de la compaction chez le matériel granulaire vibrant

La compaction vibratoire des matériels granulaires est très importante dans les applications technologiques. On a utilisé plusieurs approches pour établir un rapport entre la compaction lente et les divers effets géométriques sans faire une conclusion unique sur la nature de l'évolution temporelle de la densité du matériel. Analysant les préalables résultats expérimentaux on a pu constater que la relaxation de la densité du matériel granulaire suit la loi Mittel-Leffler dans tout le domaine temporel. On a présenté aussi le modèle fractionnel de la compaction granulaire qui est en accord avec cette loi. Dans ce travail on essaie d'identifier les directions des recherches sur les propriétés des matériels granulaires à partir des besoins d'industrie.

Mots clés: **matériels granulaires, compaction granulaire, cinétique fonctionnelle, application militaire**