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Uniaxial compression of 2d and 3d packings : electrical conductivity measurements

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Résumé. — Nous avons mesuré la conductivité d'empilements bi et tridimensionnels de grains conducteurs soumis à une pression uniaxiale verticale. La conductance en fonction de la contrainte suit une loi de puissance dont l'exposant est différent de l'exposant microscopique prévu par la loi de Hertz. Nous proposons une explication théorique simple.

Abstract. — We measured the conductivity of 2d and 3d packings of conducting grains under a uniaxial vertical pressure. The conductance as a function of stress is a power law with an exponent which is different from the microscopic Hertz exponent. We propose a simple theoretical explanation.

1. Introduction.

The properties of granular media depend generally on the geometrical properties of the system, on the properties of the grains themselves, but also on the properties of the *contacts* between grains. For the mechanical or transport properties, these contacts are very important. Our goal here is to study the effects on the electrical conductivities of the materials, of the heterogeneities induced by uniaxial pressure on packings of spheres or cylinders.

The microscopic strain-stress law at the contact between two grains may be written (Hertz law)

$$f \sim (\Delta h)^\mu \quad \text{with} \quad \mu \sim 1.5. \quad (1a)$$

Recently [1], we have verified that in a compact granular packing submitted to a uniaxial force, the strain-stress law may also be written in a « power » form

$$F \sim (\Delta H)^m. \quad (1b)$$

In general, the macroscopic exponent m is different from the microscopic one : $m \neq \mu$; it strongly depends on the material, the quality of the contacts between grains and the way the packing is made. Photoelastic studies for plexiglass cylinder packings show that the force is transmitted through a connected subnetwork of the network of the real contacts (the so-called strongest stresses network), which develops in the bulk as the stress increases and new contacts appear ; its geometry depends on the geometrical features of the packings (defects and ordering in monosize systems [1], grain size distribution in mixtures [2]...).

When the grains are conducting, the conductance g at the contact between two grains derived from Hertz microscopic law reads

$$g \sim f^\tau \quad (2)$$

where $\tau \sim 1/3$. As in the above mechanical case, at the macroscopic level, we expect a power law

$$G \sim F^t. \quad (3)$$

Such a law is surely not true, with the same exponent, on the whole range of force : at low stress, grains rearrange by rotations and local slidings (consolidation phase) and Hertz deformations surely

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plays a little role. Our experimental situation corresponds to an intermediate force range, where only elastic deformations take place and yield the main contribution to the compression. All possible contacts are not created; the strongest stresses network plays a prominent role as electrical current chooses preferentially good mechanical contacts. The resistance of a contact depends on the applied stress, first because of « Hertz effects » (increasing of the contact surface with increasing applied pressure), but also because pressure can destroy the insulating layer at the surface of the grains (oxyde, impurities, etc.); then, if it exists, the exponent t may be different from $1/3$. Finally, at large force, all contacts are realized, and one expects $t \sim 1/3$; practically, the plasticity threshold is reached long before and other processes like rupture may occur. One interesting question is the force range of the three regimes which are schematized in figure 1.

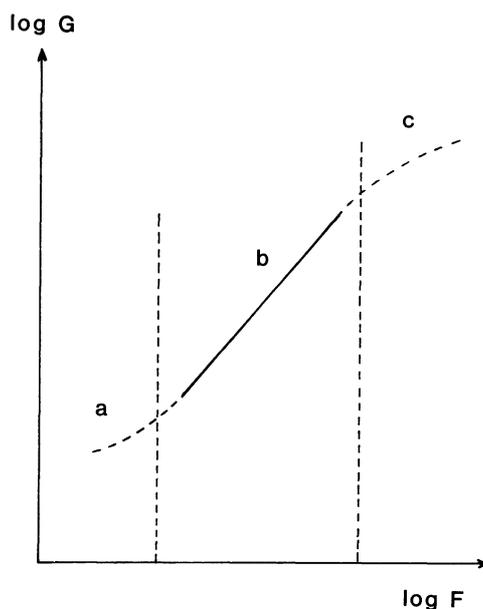


Fig. 1. — Schematic behaviour of conductivity vs. the pressure in log-log scale and arbitrary units: a) consolidation phase, b) experimental region, c) asymptotic behaviour (not reached practically).

Such a behaviour was described in a review paper by Euler [3] for compacted powders. The experimental intermediate exponent t is actually observed on one or two decades in pressure. Depending on the powder under study, t varies from $1/3$ to $5/3$, with some anomalous values ($t < 0$ or $t = 8$). However, in the cases reviewed by Euler, the process is only partially reversible as the grains remain stucked together and deformed.

We present here the results of conductivity meas-

urements of 2d (cylinders) and 3d (spheres) conducting model packings under uniaxial vertical pressure; we remain in the elasticity zone so that the compression process is reversible. In section 2, we describe the experimental apparatus and the model packings; in section 3, we give the experimental results. Finally, in section 4, we propose a theoretical model already used to explain the mechanical behaviour.

2. Description of the experiment.

The 3d samples are packings of calibrated steel spheres of diameter $\varnothing = 1$ mm. The spheres are packed in a cylindrical copper vessel, large enough to make the size effects negligible (diameter = 3.2 cm, height ~ 4 cm). The internal vertical walls are insulated with PVC and the upper and lower walls are coated with indium to prevent the deformation of the cell and improve the contacts between the spheres and the electrodes. To prevent a regular disposition of the spheres, we add along the upper and lower surfaces some larger spheres of diameter 2 mm which forbid the formation of a local order starting from the planar surfaces. We thus get a monosize disordered packing, with mean coordination number $z \sim 6-7$ [4].

To complete the study, we have realized a two dimensional packing made of identical horizontal steel cylinders (diameter $\varnothing = 4$ mm, length = 2.5 cm), with parallel axes (Schneebeli model [5]); the cylinders are ordered along a triangular regular lattice. The packing is made of 48 lines of alternatively 44 or 45 cylinders. Because it is built under gravity and the pressure is exerted vertically, the horizontal contacts play little role in the transmission of the force. One of the most important questions in that sort of experiment is that of the influence of the walls and of the size of the sample. A study of mechanical and photoelastic properties of samples with different sizes has shown that this influence is negligible for the packing we consider here [6]. Moreover, about 80-90 % of the applied pressure is transmitted at the bottom of the packing and so arching effects are weak.

The samples are placed in an INSTRON 1175 universal testing machine and a uniaxial vertical pressure is applied, growing from 0 to 4 000 N [1, 6]. The electrodes are made of copper so that their resistivity is as small as possible. Several pressure cycles are performed until the results are reproducible: 3 or 4 cycles for cylinders (the regular packing is rapidly stabilized) but 40 cycles for spheres. The measurements are done at increasing pressure as the force is no more uniaxial on the decreasing part of the hysteresis [7], and for each point, we wait until the system is in equilibrium.

The results given in section 3 for the macroscopic exponent t are averaged over three different samples.

3. Experimental results.

3.1 CONDUCTIVITY OF 3d PACKINGS. — i) In a first experiment, the metallic spheres are used as given by the manufacturer. They are « dirty » and exhibit under the microscope irregularities prejudicial to the quality of the contacts. However, the packing is conducting from the beginning and the law for conductivity vs. applied force looks as indicated in figure 1 with a transition at $F \sim 400$ N. According to the sample (essentially to the surface of the spheres), the exponent we have observed when $400 \text{ N} < F < 4\,000 \text{ N}$ (i.e. one decade in pressure) varies between 2.5 and 3.

ii) Then, the spheres are cleaned with alcohol and they are cleaned again from time to time. Under the microscope, they look clean and regular. The packing is conducting from the beginning, but the resistivity at low force is much weaker (100 or 1 000 times) than in i), which indicates that the electrical contacts are much better. The behaviour is again like in figure 1 but with a smaller intermediate exponent $t = 1.8 \pm 0.1$, valid for more than one decade in pressure (see Fig. 2a).

iii) The experiments above were done on a « large » packing, 41 mm high, which contains about 35 000 spheres. To be sure that we are in the conditions where the finite size effects are negligible, we have performed the same study with packings of lower height. The exponent remains constant and close to 1.8 even for small heights. At $h = 7$ mm (i.e. when there are about 10-12 layers of spheres) a transition occurs and t decreases to value 1.4 at $h = 3.7$ mm.

iv) The same experiment was then performed with partially ordered packings: we fill the container, layer by layer, and try to rearrange each layer in the more ordered way possible. Contrary to the previous case, the exponent t remains equal to 1.4 up to large heights, of the order of 30 mm (i.e. 50 layers). Then, it begins to grow up to the value $t = 1.8$. Because of the size of our cell, it was not possible to prepare packings higher than $h = 41$ mm; moreover, it is technically difficult to order — even partially — large packings, the fluctuations are rather important. Thus, it is not possible to decide whether we are still in the transitional domain or not. There is *a priori* no reason that the asymptotic exponent t would be the same in ordered and disordered packings.

An explanation may be given by reference to the photoelastic studies on plexiglass cylinders in the mechanical problem [1, 6]. When the height of the packing is small, strength arms go from the top to the bottom of the container and transmit most of the force (and of the current when grains are conducting). When the height increases, strength arms

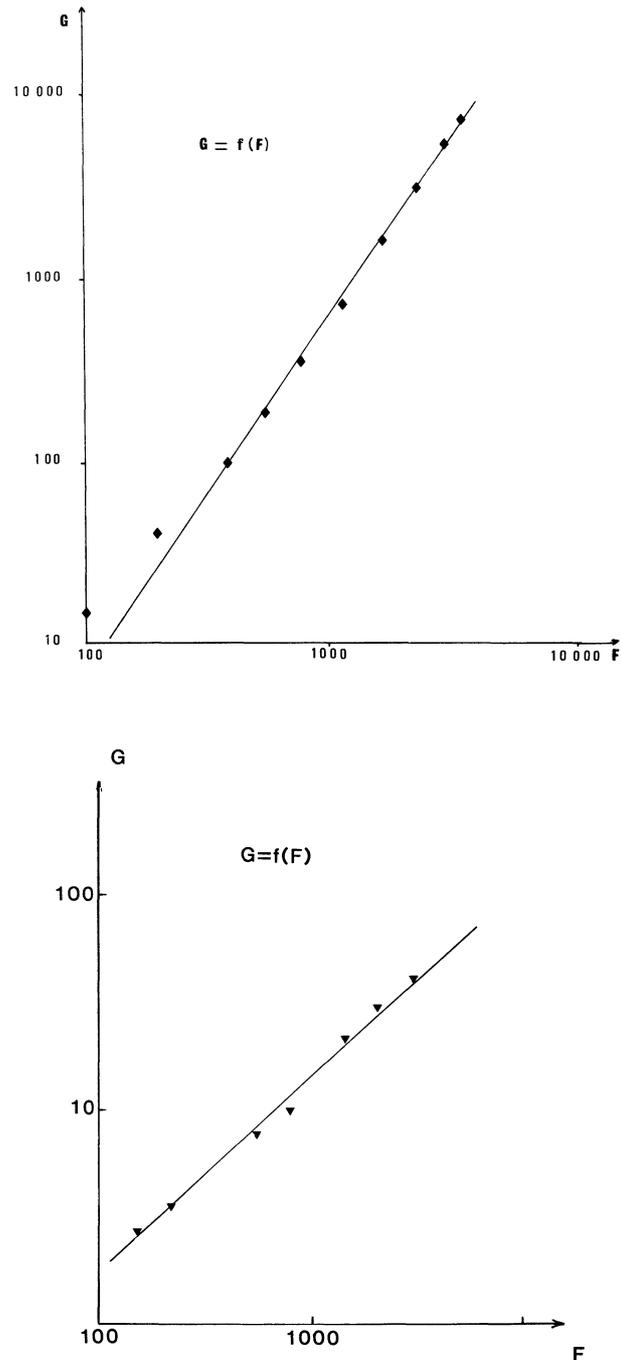


Fig. 2. — Log-log plot of the conductivity (in arbitrary units) as a function of the macroscopic force (in Newtons), a) for a disordered packing of spheres ($h = 41$ mm, 35 000 spheres), b) for an ordered packing of 1 600 cylinders; the fluctuations at high pressure become very important.

begin to interfere. In disordered systems, this behaviour occurs for small heights while in the ordered case long quasi rectilinear strength arms may exist, generated from some grains of the upper layer (orientational order seems to play an important role in that case) and this could explain the discrepancy

between the transitional heights (4 mm and 30 mm) in the disordered and ordered cases respectively.

3.2 CONDUCTIVITY OF 2d PACKINGS. — We performed a similar experiment on packings of cylinders. There is again conduction from the beginning, and in a log-log scale, the experimental points are roughly along a straight line in an intermediate zone which covers one decade in pressure (200 to 2 000 N). We get a weaker exponent

$$t \sim 0.9 \pm 0.1$$

which is nevertheless clearly distinct from 1/3 (see Fig. 2b). When $F > 2\,000$ N, the measured quantities are so small that parasite effects (for example, the electrodes are not rigorously equipotential) may become important and the experimental plots show off large fluctuations; it is not possible to test whether t is close to 1/3.

Because of the fluctuations, it was not possible to perform a size effects study like for the spheres.

4. Theoretical models.

The behaviour in « three regimes » (Fig. 1) may be understood using electrical analogies such as diodes with random thresholds on regular lattices [8], but it is not possible to use these models to describe at the same time the mechanical behaviour [9]. We propose here two simple models which describe qualitatively both mechanical and electrical exponents, provided the good mechanical contacts are too the good electrical contacts. In the first model, we use an effective medium theory, in the second one we try to take into account the connected aspect of the strongest stresses network. Their results are compatible and agree qualitatively with the experimental exponents.

In both cases, we assume that the packings behave more or less like the regular packing with the same coordination number z . *The disorder of contacts*, which leads to a variation of the total number of contacts as the applied pressure increases, seems to be more important than translational disorder [1]. The 2d cylinder packings are easily modeled from a distorted square lattice with $z = 4$ (horizontal contacts are inefficient for transmitting the force). Our 3d disordered sphere packings which have a coordination number $z \sim 6$ and a packing fraction $c \sim 0.60$ will provide intermediate values between the simple cubic ($z = 6$, $c \sim 0.52$) and the body centred cubic ($z = 8$, $c \sim 0.68$) lattices.

4.1 EFFECTIVE MEDIUM THEORY MODEL. — The percentage of good electrical contacts is an increasing function $\alpha(P)$ of applied pressure P . We assume that they are distributed at random in the sample. As far as the scale of the heterogeneities is smaller than the sample size, the effective medium theory is

believed to work and the mean conductance $g_m(P)$ at pressure P is given by [10]

$$\alpha \frac{g_0(P) - g_m(P)}{g_0(P) + \lambda g_m(P)} + (1 - \alpha) \int_0^P \frac{g_{P'}(P) - g_m(P)}{g_{P'}(P) + \lambda g_m(P)} \times N(P') dP' = \frac{1 - \alpha}{\lambda} \int_P^\infty N(P') dP' \quad (4)$$

where $\lambda = z/2 - 1$, z is the coordination number, $\alpha = \alpha(0)$ is the number of mechanical contacts which exist at $P = 0$, $g_0(P)$ is the conductance at pressure P of these contacts and $g_{P'}(P)$ the conductance at pressure P of the contacts which are created at pressure P' ; $g_0(P)$ and $g_{P'}(P)$ are given by the microscopic law equation (2). The distribution function $N(P')$ of the mechanical thresholds tells how rapidly these contacts are created. It is normalized so that

$$\int_0^\infty N(P') dP' = 1$$

and

$$\alpha(P) = \alpha + (1 - \alpha) \int_0^P N(P') dP'.$$

The microscopic force f at the contact between two grains — which is needed to calculate $g_{P'}(P)$ — is determined by assuming that the packing is arranged in layers which, on average, all behave in the same manner; in each layer, all grains, whatever stressed or not, undergo the same vertical displacement. This is the model of Ko and Scott [11] for sands; further applications and improvements may be found in reference [12].

Thus, the parameters are

i) the percentage α of contacts which are active from the beginning. One knows that it is surely rather small, of the order of 0.3 or 0.4 in the vulnerability measurements by Ottavi *et al.* [13]; it might be still smaller for badly calibrated « grains » ($\alpha = 0.15$ or 0.20 for plexiglass cylinders as seen in photoelasticity).

ii) the distribution law $N(P')$. It seems that contacts are not easily established at small force, their number increases rapidly afterwards [13]. This behaviour may be simulated with a polynomial law :

$$N(P') = \left. \begin{array}{l} \frac{k+1}{P_m} \left(\frac{P'}{P_m} \right)^k \quad P' < P_m \\ = 0 \quad P' > P_m \end{array} \right\} \quad (5)$$

where k is an integer and P_m a maximal pressure.

In table I we give the intermediate exponents t for several percentages α and coordination numbers z

Table I. — *Macroscopic intermediate exponent t for several lattices and different values of the rate α ; the distribution law for creating new contacts is given by Equation (4) with $k = 1$.*

Lattice \ α	0.3	0.4	0.5	0.7
Distorted square	—	—	2.10	1.0
Simple cubic	≥ 1.8	1.47	1.20	0.70
Body centered cubic	1.60	1.14	1.05	0.65

when $k = 1$. At very high pressure (all the contacts are created), we get the Hertz exponent 1/3. When $\alpha < 0.5$, the intermediate zone runs over one or two decades in pressure ; it is smaller when $\alpha > 0.5$, but this is probably not the experimental situation. When $\alpha < \alpha_c$, the fraction of electrically active contacts at the percolation threshold (in the effective medium model, $\alpha_c = 1/(\lambda + 1) = 2/z$), the packing is not conducting at low pressure, but it may become conducting beyond a threshold pressure P_0 where $\alpha(P_0) = \alpha_c$. This has not been observed in our packings made of conductors only but exists in mixtures of insulating and conducting grains [14]. The exponent t increases with parameter k . It may become important near the percolation threshold (if $\alpha = \alpha_c$, $t \sim k + 4/3$) ; this could give an explanation to the high exponents (2.5 to 3) found for the dirty spheres (see Sect. 3.1).

Notice that the geometrical reorganization of the consolidation phase is not well described as only Hertz deformations are taken into account : at low force, we necessarily recover exponent 1/3. It is possible to include in the model the destruction of part of the initial contacts and recreation of new contacts and so to take into account rotation and local sliding. This will be done elsewhere [12].

This model is not entirely satisfying, in spite of a correct agreement with experiment, as we know that good contacts are not at all at random and may even run along strength arms longer in ordered than in disordered systems [1].

4.2 INTRODUCTION OF THE STRONGEST STRESSES NETWORK. — It is possible to get a perhaps more realistic description in assuming that the strongest stresses network is macroscopically regular. Let us first consider 2d models. Roughly, we assume that the more stressed bonds, which are conducting from the beginning form a regular distorted square lattice (horizontal bonds are inefficient), in which each link is made of n bonds. At a threshold pressure P_1 , the strength arms are divided into two equal

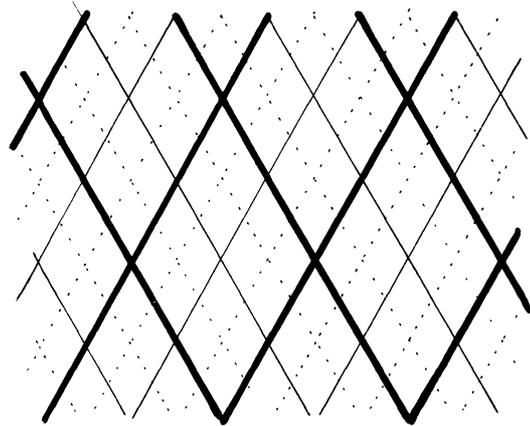


Fig. 3. — Schematization of the strongest stresses network in the 2d ordered model. a) large full line : initial step, b) thin full line : strength lines created at pressure P_1 , c) dotted line : strength lines created at pressure P_2 .

arms made of $n/2$ bonds, then at a new threshold pressure P_2 , all the arms are again divided into arms made of $n/2^2$ bonds... (see Fig. 3). In that simplistic image, the network remains a distorted square lattice. The conductance may be written :

$$G = g_0 + g_1 + 2 g_2 + 2^2 g_3 + \dots + 2^{s-1} g_s, \quad d = 2 \tag{6a}$$

where g_i is the conductance of the new arms which appear after i divisions and is given by the microscopic law equation (2). A similar relation holds for a cubic lattice :

$$G = g_0 + 3 g_1 + 3.2^2 g_2 + \dots + 3.2^{2s-2} g_s, \quad d = 3. \tag{6b}$$

Practically, for a 2d ordered packing, the number of divisions s is reasonably equal to 2 or 3. For our 3d quasi ordered packing, it may reach the value 4.

We performed some numerical tests for $s = 2, 3, 4$ and $d = 2$ or 3. We have considered two cases which correspond approximately to a constant or a linear function $N(P)$ in the previous model :

- a) new contacts set in at regular pressure intervals

$$P_i/P_m = 2^i/2^s \quad \text{if } d = 2$$

$$\text{(resp. } P_i/P_m = 2^{2i}/2^{2s} \quad \text{if } d = 3)$$

- b) new contacts set in slowly at low pressure and more rapidly at the end of the pressure interval $(0, P_m)$

$$(P_i/P_m)^2 = 2^i/2^s \quad \text{if } d = 2$$

$$\text{(resp. } (P_i/P_m)^2 = 2^{2i}/2^{2s} \quad \text{if } d = 3).$$

Actually, we have included some fluctuations around the different thresholds so that the new

contacts set in more regularly. Results are given in table II. Columns a) and b) correspond to the laws a) and b) above. The exponents increase with the number s of divisions ; their numerical values are compatible with the experiments described in section 3.

It is not easy to compare results obtained with the two models, though, in each case, the problem reduces to the estimation of the percentage of good contacts : in the last model, we deal only with the backbone of the mechanical network while in the first one, $\alpha(P)$ includes dead arms which may be important.

Table II. — *Electrical exponent in the ordered model for 2 and 3 dimensional regular packings and $s = 2, 3, 4$. Columns a) and b) correspond to a constant and a linear set in of the contacts respectively.*

s	d = 2		d = 3	
	a	b	a	b
2	0.8	1.0	0.93	1.33
3	0.95	1.1	0.97	1.55
4	1.0	1.4	1.05	1.70

5. Conclusion.

We have verified that the conductivity of a packing of steel spheres behaves as indicated in figure 1 in an intermediate zone running over one or two decades in pressure, as was already observed in the strain-stress law. The macroscopic exponent depends on the quality of the contacts and on the geometry of the packing. This behaviour may be described by simple models which work both for mechanical and electrical laws.

The experiments described above deal with packings of conducting grains only. Actually, we often need mixtures of insulating and conducting grains. The current may take place only above a minimal pressure (pressure threshold) where the rate of conducting contacts reach the critical site-bond percolation threshold. The model in section 4.1 may certainly be used to explain this situation as it corroborates, at least qualitatively, measurements made on mixtures of spheres [14]. Experiments with mixtures of equal cylinders made of steel and rubber will soon be performed to this end.

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