Experiment and Theory of the Electrical Conductivity of a Compressed Granular Metal

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Abstract. We measured the electrical resistance of a copper powder sample under uniaxial compression. The resistance is controlled by the oxide layer on grains. It follows a power law versus the pressure, with an exponent which is different from that expected either with Hertz or plastic contact between grains. A theoretical explanation based on a description of two grains in contact is proposed. We show that the strong dependence of the macroscopic resistance with the pressure applied to the powder is a consequence of large variabilities and heterogeneities present at the contact surface between two grains.

Keywords: Electrical conductivity of granular materials, Hertz or plastic contact, Electric contacts, Oxides, Lévy distributions
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INTRODUCTION

The electrical transport in metallic granular media presents interesting and astonishing properties. For example, the electrical resistance of an oxidized metallic powder which is initially high suddenly falls down by several orders of magnitude as soon as an electromagnetic wave is produced in its vicinity ("Branly effect") [1]. This instability is associated with strong 1/f noise, giant fluctuations of electrical resistance and slow relaxations [2, 3]. Notice that all theses fascinating effects only exist when the grains are oxidized. Clearly, the oxide layer that covers each grain plays a crucial role in the onset of the insulating to conducting transition.

Metallic granular materials have also found numerous applications in industry (combustion reactors, batteries, tire fabrication processes...) thanks to their unique properties which result from both the grains themselves (such as their bulk elasticity, plasticity, electrical resistivity and also the state surface), and the way they are arranged in the packing (density, state of compaction, ordered or disordered structure, existence of "force chains" which carry most of the applied stress).

Another motivation for the study of electrical transport in granular packings is the possibility to use electrical resistance as a testing tool in the investigation of force chains or force distributions. Indeed the resistance of the contact between two grains depends on the local stress so that exploring the distribution of contact resistances may give information concerning the spatially inhomogeneous force distribution in a granular media [4]. To reach this goal, it is necessary to investigate the electrical conduction through granular packings and the first step is to understand how the macroscopic electrical resistance depends on the mechanical force applied to the grains.

THEORETICAL BACKGROUND

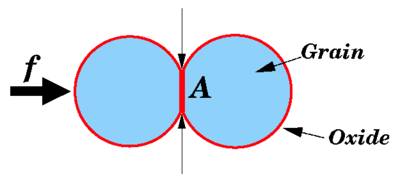

FIGURE 1. Two metallic grains under compression. A is the contact area. An insulating film composed of oxides or impurities covers each grain.

Mechanical behavior of two spherical grains in contact. The mechanical behavior of two spherical grains in direct contact is rather well known (a basic situation is shown Fig. 1). Indeed, for small static forces, the Hertz law describes remarkably well the non-linear interaction between tho elastic spheres. It predicts that the contact area (A) as a function of the force exerted on the spheres (f) follow a power-law behavior [5]:

$$A \sim f^{2/3} \qquad (1)$$

The force divided by the contact area defines the contact pressure p. Then, in the elastic regime (ER), $p \sim f^{1/3}$. At higher forces, when the pressure exceeds the yield strength of the material, permanent plastic deformation occurs at the contact and the previous power-law becomes:

$$A = f/Y \qquad (2)$$

In this plastic regime (PR), the contact pressure stays almost constant and approximately equal to the yield stress (Y) whatever the force applied to the grains.

Electrical behavior of two spherical grains in contact. Contrary to the mechanical behavior, the force dependent electrical resistance of two metallic grains in contact

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is still an open problem. For the special cases of clean and non oxidized surface contact, experimental and theoretical studies showed that the electrical resistance thus measured is proportionnal to the inverse of the cube root of the force when the contact pressure is lower than the plastic yield stress and to the inverse of the square root of the force in the plastic regime [6, 7, 8]. Indeed, the resistance of the metallic contact (r) is a result of the constriction of the current stream through the small contact area (A). It depends only on the resistivity of the metal (ρ_m) and on the radius of the contact area: $r \sim \rho_m / A^{1/2}$. For elastic spheres (Eq. 1), we get $r \sim 1/f^{1/3}$ whereas Eq. 2 (PR) leads to $r \sim 1/f^{1/2}$. However, in realistic granular packings, grains are not clean and covered by an oxide layer or another contaminant film which resistivity is largely greater than the metal one. In that case, both the bulk of the metallic grain and the constriction resistances are lower than the resistance of the insulating layer. Assuming the Ohmic law is valid and the current passes uniformly through the oxide film, the contact resistance can be written as:

$$r = \rho_{ox} \delta / A \qquad (3)$$

where ρ_{ox} and δ are respectively the oxide resistivity and its thickness. Using Eq. 1 or Eq. 2, this very simple model of the electric contact predicts a power law behavior of the force dependent resistance:

$$r \sim 1/f^{2/3} \quad \text{(ER)} \qquad (4)$$

$$r \sim 1/f \quad \text{(PR)} \qquad (5)$$

This theoretical scaling has not yet been observed in a compressed packing of metallic grains. Some experimental studies observed a power law dependence of the resistance with the force but with an exponent greater than one [9, 10]. For several granular materials, this exponent varies between 2 and 3!

In this paper, we report our results concerning the force dependent resistance of a copper powder and we propose an interpretation of our measurements.

EXPERIMENTAL SETUP

The geometrical setup of the experiment is illustrated in Fig. 2. The granular material is a copper powder composed of roughly spherical particles with a diameter included between 50 μm and 110 μm [11, 3, 12, 13]. Two grams of powder are confined in a plexiglass cylinder (15 mm of inner diameter), capped with two metallic electrodes. A mechanical pressure, P, rising up to 30 N/mm^2 is applied to the powder, and is measured with a static force sensor (FGP Instrumentation™). As the packing fraction is about 0.6, an average particle diameter of

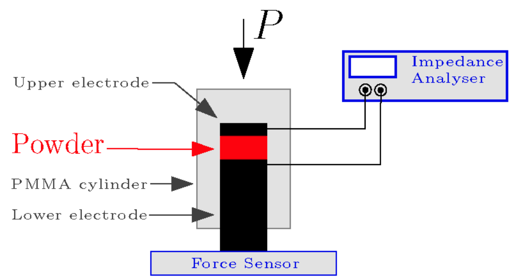

FIGURE 2. Sketch of the experimental principle.

$<\Phi>= 80$ μm leads to a density of $n = 2200$ particles per cubic millimeter. It is not easy to exactly calculate the average compression force ($<f>$) between two neighborhood grains in contact. However, we can estimate it as the product of the macroscopic pressure applied to the powder (P) with the average microscopic surface that the two grains in contact occupies ($(n \times 2 <\Phi>)^{-1}$):

$$<f>= \frac{P}{2n\Phi} \qquad (6)$$

As shown in Fig. 3, the minimum pressure applied to the powder is 1 N/mm^2. Thus, for that macroscopic pressure, the average microscopic force is 3 mN (Eq. 6). It is already enough to deform plastically the grains. Indeed, the yield stress of copper is equal to 70 N/mm^2 and, in the plastic regime, the contact area is given by Eq. 2, so that the diameter of the contact area goes up to approximately 8 μm (say $<\Phi>/10$) when the force reaches 3 mN. It is thus reasonable to think that a large number of grains in the packing endures irreversible plastic deformation. That is why the container is refilled with a new sample of powder before each experimental run.

The electrical resistance is measured using a Hewlett Packard 4192A Impedance Analyzer. Notice that the amplitude of the alternative voltage applied to the powder sample is fixed to a low value ($U_{RMS} = 100$ mV) in order to avoid any voltage-induced nonlinear effect [12, 14]. Recently, we showed that the resistance is frequency independent at low frequencies (< 1 kHz) [12, 13]. In addition, the ac conductivity collapses to the dc conductivity when the frequency is low. The main interest to use an impedance analyser in place of a dc multimeter is the possibility to measure both the amplitude and the phase of the electrical response of the powder. Indeed two metallic grains in contact by an insulating layer (Fig. 1) can be electrically described as a resistor and a capacitor connected in parallel. Thus, at a macroscale, the complex impedance of a powder sample is modeled by a parallel resistance-capacitance combination. Measuring the capacitance gives us additional information about the electrical contact. The simple model described in Sect. "Theoretical Background" (see also Fig. 1) predicts that the capacitance (c) of the contact between two grains under compression can be written as:

$$c = \varepsilon A/\delta \tag{7}$$

where ε and δ are respectively the dielectric permittivity and the thickness of the oxide layer. Combining Eq. 7 with Eq. 4 or Eq. 5, a power law behavior of the force dependent capacitance is obtained:

$$c \sim f^{2/3} \quad \text{(ER)} \tag{8}$$

$$c \sim f \quad \text{(PR)} \tag{9}$$

Next section we give the comparison between thoses power law relations and our experimental results.

RESULTS

In Fig. 3 we plot the electrical resistance as a function of the applied pressure. In the pressure range from 1 to 20

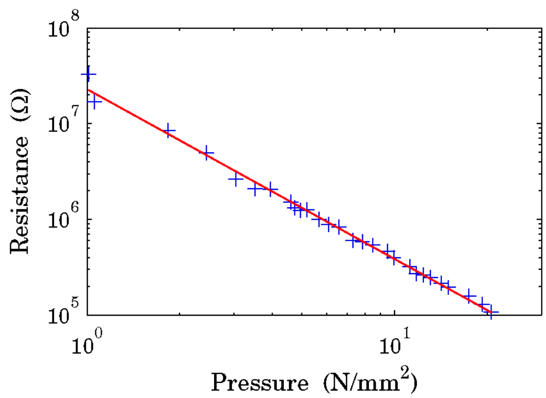

FIGURE 3. Electrical resistance as a function of applied pressure. The line is power-law fit to the points shown. The exponent is -1.8.

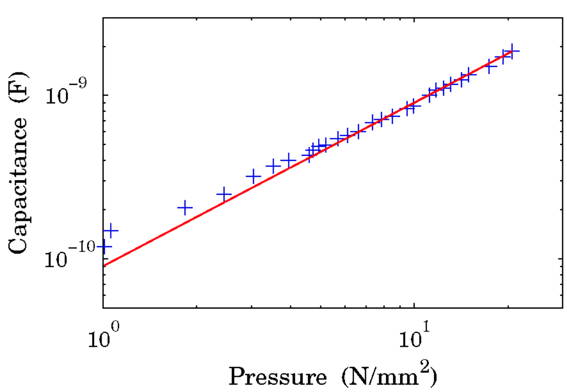

FIGURE 4. Capacitance vs pressure. The frequency is fixed at 20 Hz. The line is power-law fit with slope equal to one.

N/mm^2, the resistance (R) seems to follow a power law behaviour like:

$$R \sim P^{-\alpha} \tag{10}$$

where $\alpha \approx 1.8$.

The capacitance as a function of pressure exhibits also a power law scaling (see Fig. 4). We find that the capacitance is proportionnal to the pressure, in excellent agreement with the prediction from Eq. 9.

Thoses experimental results concerning the capacitance confirm the plastic behavior of the grains (Eq. 2) and also the validity of the simple electrical model in where an insulating layer is taken in sandwich between two metallic grains (see Fig. 1). On the contrary, the resistance displays more complicated features since it decreases more rapidly than the inverse of the pressure (Eq. 10). Clearly, there is a significant difference of behavior between the resistance and the capacitance pressure dependences. Whatever the mechanical regime (elastic or plastic) that describes the contact, our simple model (Eqs. 4 or 5) fails to predict an exponent α greater than one. Next we propose a new model for the electric contact that explains well why the resistance and the capacitance behave so differently.

A NEW MODEL FOR THE ELECTRIC CONTACT BETWEEN TWO GRAINS

To understand why the resistance decreases faster than the inverse of the pressure, let us modify the electrical model of two grains in contact described in Sect. "Theoretical Background" (Eq. 3). Maybe, the insulating layer is a non homogeneous material since there are numerous different oxides or impurities not uniformly distributed at the surface of each grain. Instead of passing uniformly through the oxide layer, we assume that the electric current prefers to be divided in a large number of microcurrents following conducting microchannels [15]. The conductivity of thoses microchannels is greater than that of the insulating material around them because they are constituted of good conducting oxides plus probably some atoms of copper coming from the metallic grain. The number of available conducting microchannels (N) obviously increases with increasing the contact area. Besides, assuming N proportionnal to A is not a restrictive assumption. Thus, in the plastic regime:

$$N \sim A \sim f \tag{11}$$

Since the microchannels of conduction are connected in parallel, the total contact conductance (Γ) is expressed as the sum of their individual conductance (γ_i):

$$\Gamma(f) = \sum_{i=1}^{N(f)} \gamma_i \tag{12}$$

As the number of microchannels is large, we can adopt a statistical description of their conductance. We define Q the distribution of γ_i. To each microchannel i, we asign a conductance $\gamma_i > \gamma_0$ drawn at random from Q. Below γ_0, the resistance of the microchannel is too high and no current flows through it. Q crucially depends on the physical properties of the oxide layer. A large variability of the characteristics of the microchannels (nature of the oxide, length, width) leads to a broad distribution of γ. Hereafter, we assume that $Q(\gamma)$ does not show any significant force dependence that is to say the characteristics of the oxide layer does not change when the force is increased. Only the area of contact (A) increases with increasing the mechanical force.

In the case of a sufficiently large number of conductances and also finite mean ($<\gamma>$) and finite variance for $Q(\gamma)$, the Central Limit Theorem states that the sum of N independent conductances tends to $N\times <\gamma>$. Then: $\Gamma \sim N \sim A$. The contact resistance ($r = 1/\Gamma$) is thus proportionnal to the inverse of the contact area so that the law obtained for an uniform oxide layer (Eq. 3) is recovered. In the plastic regime, it yields to a resistance proportionnal to the force.

In the case of very broad distribution, that is to say if there is huge variability in the conductance of the microchannels, the average conductance may not exist. Let us give an example. Suppose that the distribution Q decays according to a power-law:

$$Q(\gamma) = \mu \frac{\gamma_0^\mu}{\gamma^{1+\mu}}, \tag{13}$$

the parameter μ controlling the tail of Q. Large values of μ give narrow distributions of γ whereas values of μ close to zero give broader distributions. The degree of disorder and the variability increase when μ decreases. Hereafter, $0 < \mu < 1$ so that $<\gamma>$ does not exist. The standard Central Limit Theorem does not apply, but its generalization states that the sum of N independent conductances tends to a N-independent Lévy distribution Ψ [16, 17]:

$$\Gamma \sim N^{1/\mu}\Psi \tag{14}$$

Note that $1/\mu > 1$. When the force increases, both the contact area and the number of microchannels increases proportionnally to the force ($N \sim A \sim f$ in the plastic regime), but the total conductance of the contact increases faster than the force: $\Gamma \sim f^{1/\mu}$.

Finally, thanks to this example, we show that a broad distribution of the conductance of the microchannels yields to a contact resitance decreases faster than the inverse of the force:

$$r \sim 1/N^{1/\mu} \tag{15}$$

As for the capacitance, the existence of conducting microchannels does not really modify the global dielectric permittivity of the oxide layer. So, Eq. 9 stays true.

CONCLUSION

The idea that disorder and heterogeneity govern the behavior of granular packings is not recent. But there are different classes of disorder and it is a great chalenge to relate clearly each physical property of granular media to the source of disorder it comes from [18]. Here, we showed that the power law dependence of the electrical resistance with the stress can be the result of the local disorder at the contact area between two grains. It should be noted that our model only describes one single electrical contact but it reproduces well the pressure dependent resistance of the whole powder. The next challenge is to understand why such model is sufficient to describe the electrical behavior of a packing of roughly one million grains.

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