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Limit current density in 2D metallic granular packings

S. Dorbolo[a], M. Ausloos[b], and N. Vandewalle

GRASP, Institut de Physique B5, Université de Liège, 4000 Liège, Belgium

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Abstract. The electrical properties of a 2D packed metallic pentagons have been studied. The electrical characterization of such metallic pentagon heaps, like $i-V$ measurements, has been achieved. Two distinct regimes have been shown. They are separated by a transition line along which the system exhibits a memory effect behavior due to the irreversible improvement of electrical contacts between pentagons (hot spots). A limit current density has been found.

PACS. 81.05.Rm Porous materials, granular materials – 45.70.-n Granular systems – 05.60.-k Transport processes

1 Introduction

At the end of the 19th century, Calzecchi-Onesti, and then Branly, proved that a metallic packing can present both an insulating state and a conducting state [1,2]. This behavior, at the origin of radio telecommunication, is not yet understood.

Thermal analysis by Vandembroucq *et al.* [3] has shown that the Calzecchi-Onesti transition is due to the soldering of grains. The authors have observed a strong increase of the temperature along given electrical paths. This indicates welding of beads at their contacts. The welded contacts become much more conductive than the others and an insulating/conducting transition occurs when the first percolative welded path is set. Moreover Bonamy [4] has found that the current-voltage $i-V$ curve of one contact between two beads is continuous: no sharp transition occurs. Very recently, we measured the $i-V$ diagram of a 3D lead bead heap. The voltage has been found to vary exponentially with respect to the current followed by a sharp Calzecchi-Onesti transition as soon as a critical current is reached [5]. Similar effect are also shown in 2D heap packing (Fig. 1). Such a packing cannot form a compact 2D.

This paper concerns further investigation of the $i-V$ diagram of a 2D metallic grains packing before any Calzecchi-Onesti transition. For that purpose, a simple system has been considered. It consists in a packing of 1 mm thick planar pentagons for which edges are 10 mm wide (Fig. 1). Such a 2D system has been studied in references [6,7] as far as compaction properties are concerned. The impossibility for pentagons to be packed into in a compact configuration, whence form intrinsic arches, is thus very interesting to study.

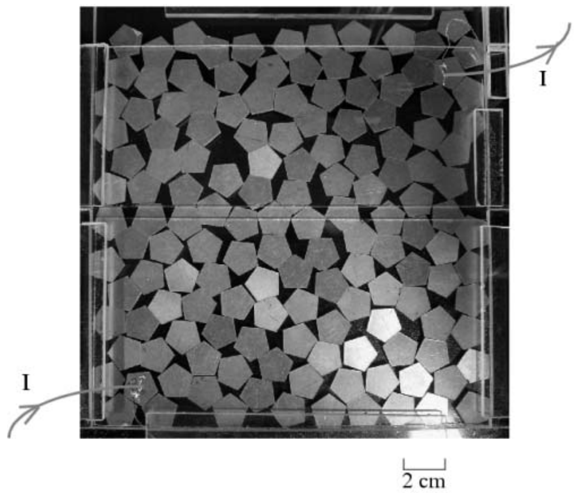

Fig. 1. The 2D-like setup containing the metallic pentagons with current leads.

We report the analysis of the voltage behaviour during current cycles. This allows us to describe the $i-V$ plane as supporting two distinct regimes. Contrary to bulk materials, granular packing is not only represented by one single curve in the $i-V$ plane but by several trajectories depending on the history of the injected currents in the system. Memory effects are thus pointed out. The points of the $i-V$ plane define an area limited by the so-called irreversibility line and the intrinsic resistance line. The irreversibility line is the line along which the electrical resistance is weakened when the current increases.

[a] e-mail: `s.dorbolo@ulg.ac.be`
[b] Also at SUPRATECS

In Section 2, the experimental setup and conditions are described. The next two sections are devoted to results and interpretations. Conclusions are drawn in Section 5.

2 Experimental setup

About 200 aluminium pentagons are placed in a plexiglas vertical vessel of 210×200 mm^2. In order to avoid overlaps between pentagons and, in so doing, to keep 2D conditions, the vessel width is 1 mm thick (Fig. 1). The packing is obtained by randomly dropping the pentagons between the two vertical planes. No further compaction process has been performed. The electrical contacts are set on two particular pentagons. These contacts are soldered and the measurement of the resistance is performed in a two-wires configuration: the lowest measured global resistance of the packing is about 20 Ω which is much higher than the one of the current leads.

In order to avoid electromagnetic perturbations, the vessel and the measurement instruments are placed in a Faraday cage. Electrical contacts are connected by coaxial cables to a Keithley K2400 current source which allows also to measure the voltage. During the experiment, the current intensity i was imposed and the voltage V measured.

The injected current is successively set to i = sign 10^{-q} A, q being the running parameter in the following description. A maximum current value is set by fixing q_{max}. Starting from the minimum current $q = 6$, the current is increased up to $q = q_{max}$. It is then decreased to $q = 6$ and to "negative" values. To summarize, a cycle is defined when q varies as follows

$$\begin{aligned} \text{sign} = 1: \; & q = 6 \rightarrow q_{max} \\ & q = q_{max} \rightarrow 6 \\ \text{sign} = -1: \; & q = 6 \rightarrow q_{max} \\ & q = q_{max} \rightarrow 6. \end{aligned} \tag{1}$$

A new value for q_{max} is fixed and another cycle is made. The maximum values have been successively set to 4, 3, 2.5, 2, 1.5 and finally 1 which corresponds to the maximum current intensity that the source can deliver (100 mA).

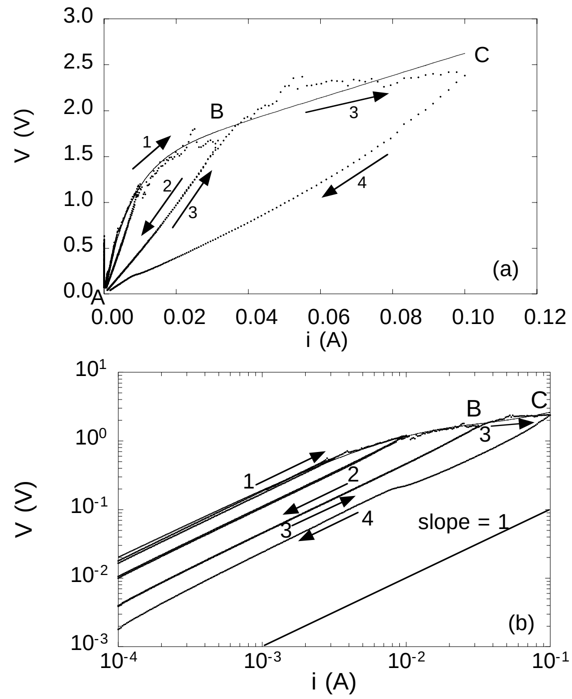

Fig. 2. (a) The $i - V$ curve obtained for a cycle of injected currents as described in the text, in the upper positive quadrant values. Numbered arrows indicate the different stages of the measurement. A, B and C are particular points discussed in the text. (b) Same results present in a log-log plot in order to better emphasize the different cycles.

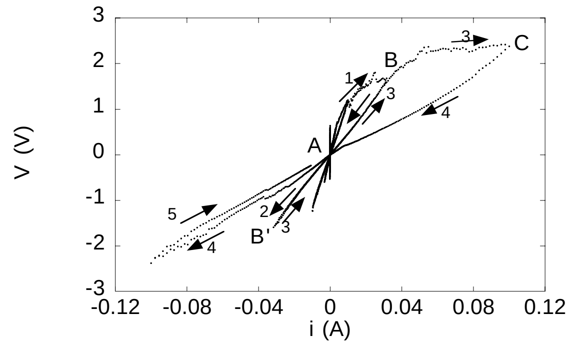

Fig. 3. The $i - V$ curves obtained during successive current cycles of injected currents up to $i_{max} = \text{sign}10^{-q}$ value.

3 Results

The recorded $i - V$ curves are shown in Figure 3. Since the range of current intensities is broad, the first quadrant data has been presented in Figure 2a and in a log-log plot in Figure 2b. Figure 3 is enlarged in Figure 4 to show the measurements obtained during the first loop at small i, *i.e.* $q_{max} = 4$.

In order to describe a cycle, let us follow the arrows indicated on Figures 2a, 2b and 3. The point A denotes the starting point. First, the current is increased. The voltage is seen to increase according to the law (materialized by a solid line in Fig. 2a and b)

$$V = [V_0(1 - \exp(-i/i_0))] + R_r i \tag{2}$$

where R_r the intrinsic electrical resistance of the packing. The system reaches point B, for example for $q_{max} = 1.5$. The current is then decreased (arrow 2); the voltage decreases following a *linear* law $V = R_a i$ where R_a is a fitting coefficient, until point B' (Fig. 3). Next, the current is increased until 10^{-1} A (arrows 3). The voltage first follows the same linear law till point B. After that, it follows the law of equation (2) till point C. The injected current i is then decreased (arrows 4): the voltage linearly decreases as $V = R'_a i$, where R'_a is *lower* than R_a.

A close look at Figure 3 shows that the feature still exists for low injected currents at the beginning of the experiment. This structure is emphasized in Figure 4. The

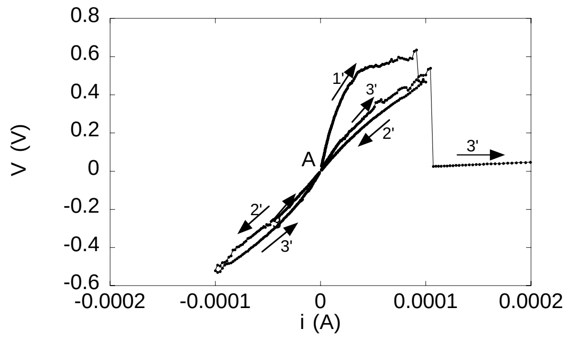

Fig. 4. The $i - V$ curve as in Figure 3 for the first very low injected currents cycles.

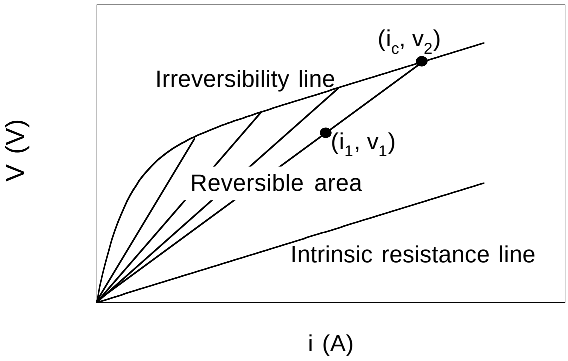

Fig. 5. Sketch of the different zones and curves presented in the $i - V$ diagram.

initial electrical resistance is very high: about 25 kΩ; while the structure of the $i - V$ curve is similar to that of Figure 2a. This feature would not be observed for injected current higher than the ones of this example. That shows the high hysteretic behavior of such a system and the existence of different scales are in presence in a granular packing. The jump at $i = 10^{-4}$ A is attributed to a breakdown of the reverse diode as observed in a 3D system [5]. This breakdown will be discussed herebelow.

4 Interpretations

In order to summarize our results, Figure 5 is a sketch of Figure 2a. The irreversibility line is described by equation (3). Microsoldering processes are assumed to occur along this line since electrical arcs can be seen during the experiment. The intrinsic resistance line is obtained when the contacts can be neglected. The resistance of a aluminium wire, representative of the soldered grains, would be then obtained. These two lines define the accessible points for the system. Ohm's law is of application as indicated by the slope of straight lines in Figure 2b.

Let us consider any point of coordinates (i_1,v_1) included in this zone (Fig. 5). The straight line which links this point to the origin is given by $V = (v_1/i_1)i$ (Ohm's law). This line intersects the curve of equation (2) at (i_c,v_2). The point (i_1,v_1) can be thus reached by increasing the current following the irreversibility line to the point (i_c,v_2) and then by decreasing the current following $V = (v_1/i_1)i$. That also means that the Ohm law is applicable in the range of the injected currents $[-i_c,i_c]$. In this range, the electrical resistance behaves as a conducting bulk material.

The different behaviors find their origin in the granular nature of the media. A large number of publications concerns the modelization of contact networks. They consider either a resistor network [10–15] or diode network [16,17] or mixing resistor and diode network [18,19]. Some of these models may capture the dielectric breakdown as well as the non-linear behavior of the electric resistance. Considering the chemical composition and the geometry of a contact, *i.e.* an oxide layer between two metallic pentagons, the non linearity of the electrical resistance can be introduced by considering a granular packing as a network of diodes. A contact is then represented by two opposite diodes in parallel. Moreover irreversible processes should be introduced in the model in order to account for the experimentally observed memory effect.

We propose to model contacts as if they are made by two opposite diodes in parallel plus a resistor R in parallel. Diodes have a behavior described by equation (2).

Considering the system as a continuous medium of resistivity ρ, one can write its resistance as $R = \rho\ell/S$ where ℓ is the effective length of an electrical path between the electrodes and S is the effective section of the path. When V tends to V_0, it can be rewritten $V_0 = \rho\ell i/S$. The ratio i/S equals to the conductivity σ multiplied by the limit electrical field $E_0 = V_0/\ell$ (about 5 V/m). In other words, i/S remains equal to J_ℓ which is hereby called a limit current density. This quantity remains constant in the range of high currents. This means that an increase of current intensity corresponds to an increase of the section of the contact resistor so as to keep J_ℓ constant. The grains knit together following the rule $S = J_\ell/i$. Within this approximation, the section increases by a factor 3 between the point B and C in Figure 2a. This microsoldering process is irreversible. When the current intensity decreases, the current follows the path defined by those enhanced contacts which shunts the diodes. The Ohm's law is from then on application.

The very low current behaviors shown in Figure 4 are described qualitatively by the model hereabove. When the same arguments are applied to this, a smaller J_ℓ is found since $E_0 = 1.2$ V/m in this case. Nevertheless, a breakdown occurs at 10^{-4} A (Fig. 4). This is similar to the Calzecchi-Onesti transition found in a 3D system as in reference [5]. It can be explained as a Zener or avalanche breakdown of the reverse diode when the voltage across a contact reaches the critical value, *i.e.* the Calzecchi-Onesti transition.

5 Conclusions

Current cycles have been applied to a 2D metallic pentagon packing. The measurement of the voltage during these loops shows that the strong influence of the electrical history of the packing. Moreover, non linear behaviors have been obtained and irreversible processes as microsoldering occur.

In order to take into account these different experimental facts, a weak electrical contact in granular metallic packings can be modelled as three components in parallel: two opposite diodes and a resistor. This resistor modelizes the microsoldering and is characterized by a limit current density. When this limit density is reached, the equivalent section of the resistor increases, the grains weld together. This process is responsible for the hysteretic loops in $i-V$ curves.

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