Aging process of electrical contacts 
in granular matter

S.Dorbolo,¹ M.Ausloos,¹ N.Vandewalle,¹ M. Houssa²

¹ GRASP, Institut de Physique B5, Université de Liège, 
B-4000 Liège, Belgium
² L2MP, Bat. IRPHE 
49, rue Joliot Curie 
BP 146 Technopôle de Château - Gombert 
13384 Marseille Cedex 13, France

March 22, 2022

Abstract

The electrical resistance decay of a metallic granular packing has been measured as a function of time. This measurement gives information about the size of the conducting cluster formed by the well connected grains. Several regimes have been encountered. Chronologically, the first one concerns the growth of the conducting cluster and is identified to belong to diffusion processes through a stretched exponential behavior. The relaxation time is found to be simply related to the initial injected power. This regime is followed by a reorganisation process due to thermal dilatation. For the long term behavior of the decay, an aging process occurs and enhances the electrical contacts between grains through microsoldering.
1 Introduction

The structure of granular materials is characterized by arches that distribute the weight of the packing towards the edges of the vessel. This induces an anisotropic distribution of pressures at the points of contacts between grains. The electrical resistance \( R \) of one contact has been found to depend on the applied pressure, as a power law as demonstrated by Hertz [1]. Any variations of the structure of the heap, whatever its nature (geometrical, chemical, electrical and mechanical), changes the contact network and in particular the overall electrical resistance, indeed in a way very sensitive to external perturbations [2, 3]. The information contained in \( R \) is also linked to the nature of the contacts. The surface of metallic grains may often be covered by an oxide which introduces some semiconducting properties to the contacts. This chemical layer in fact constitutes a tunneling barrier to the current flow. Therefore a granular packing may be represented by a skeleton of electrical links between grains, each link being characterized by a certain electrical resistance determined by the pressure between beads (given by Hertz relation) [4, 5, 6]. The electrical resistance can thus be a tool which allows to explore arching pressure distribution without markedly changing the structure. Such a tool can be useful in probing stress in granular system as they are submitted to a shear for example. Moreover the study of the electrical properties of granular material can give information concerning large electrical complex network.

Recent studies have shown that the electrical properties of metallic granular packings are complex, e.g. the voltage \( V \) across a sample is not univocally defined by the injected current \( i \). Indeed, current-voltage \( i-V \) curves are non-linear, sometimes singular or even discontinuous and exhibit memory effects [7, 8, 9]. A typical behavior is shown in Fig. 1. The role of the grain oxide layer has been pointed out to be responsible for the non-linear behavior of the electrical resistance with respect to the injected current [9]. Moreover taking into account the structure of the network and the nature of the contacts, the shape of the \( i-V \) curves and its discontinuities (Fig.1) may be explained [9]. The sharp transitions, first discovered by Calzecchi-Onesti [3], occur when the current reaches a certain value at which the resistance falls several orders of magnitude. This process is very similar to breakdowns in semiconducting devices [10]. This dielectric breakdown implies high local electronic reactivity, increase in temperature and grain soldering. However, microsolderings
between grains may occur before a Calzecchi-Onesti transition [11], just like nucleation sites are initiated by high concentration gradients at a first order phase transition. Those solderings are non equilibrium, or irreversible processes thereby being the fundamental cause for the hysteretic behavior found in $i - V$ curves of granular packing.

We have recorded the natural evolution of the electrical resistance of a lead beads heap versus time when a stable current flows through the system. A fixed current is injected during 20 min and the voltage is sampled every 0.2 s. Different current intensities have been injected in order to compare their effect on the variations of the resistance with time. We emphasize that this situation differs drastically from a classical $i - V$ curve measurement since in this latter case, the steps for increasing the injected current are very small and their succession very slow. The voltage has been found to decrease by about 30% depending on the value of the injected current.

The experimental set up is described in Sect.2. A discussion is found in sect.3. The conclusions are drawn in Sect.4.

2 Experimental set up

The experimental set up is simple but the different operations have to be performed with care because of the high sensitivity of the system to external perturbations. The lead beads have a mean diameter of 2.35 mm with a polydispersity of 2%. Energy Dispersive X-ray measurements have shown that the surface is made of a nanometric layer of lead oxide. About 14,000 beads have been stacked into a $50 \times 50 \times 40 \text{ mm}^3$ plexiglas parallelepipedic vessel. The density of such a 3D packing is about 0.75 and is naturally near the close packing configuration. Electrical current leads are two lead rectangular plates placed on two opposite faces of the vessel and connected to a Keithley K2400 current source [9]. The voltage $V$ has been recorded at a 5 Hz frequency on a computer. The whole system was placed in a climatized Faraday cage at 16°C; no vibration was expected to occur during the experiments.
3 Results and interpretations

One of the typical $i - V$ behavior of the lead bead heap has been reported in Fig.1. Different features which have been discussed above are indicated in the figure, i.e. (i) hysteretic loops due to microsoldering, (ii) Calzecchi-Onesti transition and (iii) a peak indicating the sensitivity of the electrical resistance to mechanical shocks.

Figure 2 presents $R(t)$ results obtained for different injected currents between 10 and 65 mA. The resistance $R$ has been obtained by dividing the measured voltage $V$ by the injected current $i$. The different lines smoothly link the experimental data points. Symbols are placed every 50 points for trend emphasis.

The resistance decreases with time whatever the injected current is. This strongly resembles the behavior obtained in the case of the degradation of ultrathin oxide layers in presence of a electrical stress. Indeed such a behavior of the voltage can be found in ref. [10, 12]. Nanolayers of insulator stressed by a voltage are studied in those works. Leakage currents induce damage to the insulator that allows more and more current to pass through. In the present case, the thin oxide layer that covers the beads is responsible for such a behavior.

The initial resistance differs according to the injected current. The higher the injected current, the lower the electrical resistance. This matches with the non-linear behavior observed in the $i - V$ curves in Fig.1 where the slope is proportional to the differential resistance. During the first 2000 seconds of observation the resistance keeps on decreasing towards some saturating value.

The evolution of the resistance for an injected current of 65 mA is particular: a Calzecchi-Onesti transition occurs after 250 s, i.e. the resistance falls by one order of magnitude from 1800 $\Omega$ to 35 $\Omega$. In this case, two processes are noticed: (i) a slow process which governs the decay of the resistance with time and (ii) an avalanche process which induces the Calzecchi-Onesti transition (like a Zener breakdown).

In Fig.3, the voltage $V$ normalized to the initial voltage $V_0 = V(t = 0)$ has been represented versus time in a semi-log plot for four different currents ($i < 50$ mA), as indicated by the legend (a normalized voltage is equivalent to a normalized resistance). The decay amplitude is larger when the current is low, e.g. for $i = 10$ mA, the decay is about 25%. Chronologically, the voltage
decay may be decomposed into three phases. (i) During the first minute, the
slope of the R-decay is about the same whatever the current value is. (ii)
The curves obtained for \( i = 10 \) and 25 mA seem to regularly decrease. The
curves obtained for \( i = 35 \) and 50 mA stabilise around \( V/V_0 = 0.82 \) during
1000 s before decreasing again. (iii) The slope of the curves are found to
be the same from the 1000th second to the end of the experiment. We may
conclude that various processes occur.

To sum up, the decay process goes as follows: (i) a primary voltage
decrease during the first minutes, (ii) a temporisation stage (due to the com-
petition of two kinds of effects), especially seen in curves \( i = 35 \) and 50 mA
and (iii) a secondary decay phase.

In Fig.4, the data are analyzed during the first phase. A stretched expo-
nential fit,

\[
\frac{V}{V_0} = (1 - a) + a e^{-\sqrt{t/\tau}}
\]  

is found where \( a \) and \( \tau \) are fitting parameters (see inset). The square root
exponent characterizes a diffusion process as in crystal growth process [13,
14, 15]. In the present case, this exponent can be interpreted as follows: The
conduction of the current through the heap is thought to be done through a
cluster of ‘well’ connected beads. If such privileged contacts exist, they shunt
the remain of the packing. Recall that electrical stresses are concentrated
along this cluster path. This means that any enhancement of other contacts
allows the connected cluster to grow as in an avalanche process. This process
is related to a diffusion of stresses through the sample. Pennetta et al. show
that such an avalanche-like propagation exists in semiconductor integreation
technology. Some percolation models have been developped. However, the
resistance decrease seems to be smoother than the one found in percolation
models [16].

The cluster growth is limited by the size of the vessel; this can explain a
saturation process. This first stage is shortened as the current is increased.
The caracteristic time \( \tau \) depends on the value of the initial injected power
\( P = V_0 i \) as \( \tau \propto P^{-1.18} \) (see inset Fig. 4). The exact value of the exponent
is quite influenced by the error bar on \( V_0 \) since this latter parameter is not
very precise and is found to change very quickly near \( t = 0 \).

The temporisation stage can be attributed to the increase of tempera-
ture in the system. Indeed, a thermocouple has measured that the inner
temperature of the heap can increase by about 20 K, depending on the injected current value. This effect is particularly noticeable for high currents, since more power has to be dissipated. The power dissipated (calculated at $t = 2200$ s) by the beads has been found to be 0.3, 1.9, 3.4 and 5.2 W for $i = 10, 25, 35$ and 50 mA respectively. Note that the local change in temperature not only modifies the nature of contacts, but also the structure of the network because of dilatation processes that may lead to a reorganization of system structure. This may cause to a further increase in the resistance since microsolderings may be damaged.

As seen before, the first decay seems to essentially concern the enhancement of the conductance due to the growth of the conducting network. On the other hand, the long term decay seems to be related to the enhancement of the contacts themselves. The thin insulating layer deteriorate with leakage current [10]. Microsolderings occur and the global resistance decreases. The system can be easily reset to its initial resistance by a tap on the vessel.

4 Conclusions

The above evidences a diffusion dynamic has been evidenced. When a current is injected through granular packing, it takes much time (more than 2000 s) for the system to stabilize its resistance. First the resistance decreases due to a network effect: the well connected cluster grows. Thus, because of the power dissipation, the temperature change induces dilatation of certain beads: the resistance may increase according to the value of the power. Finally, contacts are enhanced by the slow deterioration of the oxide layer of beads.

To summarize, the behavior of such material is thus completely different from a metallic bulk material. The perfect example is what happens in a lamp. The lamp filament has a resistance of about 50 Ω for low current and at room temperature. When the current is switched on, the lamp produces light and heat. This change in temperature makes the electrical resistance increase. On the other hand, in the case of granular material, the opposite effect occurs. When the current is first set, the resistance is high. The heat, the aging and some Zener effects affect the resistance by decreasing it. Those effects contribute to the growing of the connected cluster of beads.

Practically, as soon as some material presents weak links, electrical measurements have to be carefully achieved. The injected current history and
the injecting rate of the current are both seen to be very relevant parameters. Further theoretical works will be developed on the basis of those observations in order to model such complex electrical networks.

5 Acknowledgements

S.D. would like to thank FNRS for financial support. This work has been also supported by the contract ARC 02/07-293. We would like to thank Prof. H.W. Vanderschueren for the use of MIEL facilities. We want also to thank Dr. A. Béssinck (U. of Liège) for his valuable comments. Fruitful discussions with Prof. G. Albinet and Dr. L. Raymond are gratefully acknowledged.
References

[1] H. Hertz, J. Reine und Angewandte Mathematik 92, 156 (1895).

[2] E. Branly, C. R. Acad. Sci. Paris 111, 785 (1890).

[3] T. Calzecchi-Onesti, Il Nuovo Cimento 16, 58 (1884).

[4] S. Kirkpatrick, Rev. Mod. Phys. 45, 574 (1973).

[5] H. Takayasu, Phys. Rev. Lett. 54, 1099 (1985).

[6] S. Roux and H.J. Herrmann, Europhys. Lett. 4, 1227 (1987).

[7] D. Vandembroucq, A.C. Boccara, and S. Roux, J. Phys. III 7, 303 (1997).

[8] D. Bonamy, Phénomènes Collectifs dans les matériaux granulaires, (Ph.D. Thesis, Paris, 2000).

[9] S. Dorbolo, M. Ausloos, and N. Vandewalle, Appl. Phys. Lett. 81, 936 (2002).

[10] M. Houssa, T. Nigam, P.W. Mertens, and M.M. Heyns, Appl. Phys. Lett. 73, 1 (1998).

[11] S. Dorbolo, M. Ausloos, and N. Vandewalle, e-print cond-mat/0302288.

[12] C. Pennetta, L. Reggiani, Gy. Trefán, Physica B 314, 400 (2002).

[13] M. Avrami, J. Chem. Phys. 8, 212 (1940).

[14] A. Gadomski, Phil. Mag. Lett. 70, 335 (1994); A. Gadomski, Nonlinear Phenomena in complex systems 3, 321 (2000).

[15] P. Papon, J. Leblond, and P.H.E. Meijer, Physique des transitions de phases, (Dunod, Paris, 1999) p.52.

[16] C. Pennetta, Z. Gingl, L.B. Kiss, L. Reggiani, M. De Vittorio, A. Cola and M. Mazzer, Microelectron. Reliab. 38, 249 (1998).
Figure Captions

**Figure 1** Sketch of the $i - V$ behavior of a metallic granular material

**Figure 2** Evolution of the electrical resistance of a lead beads heap with respect to time. The different curves have been obtained for several fixed injected currents as indicated by the legend. The symbols are placed every 50 points

**Figure 3** Normalized voltage versus time obtained for different currents (see legend). One point out of 50 is indicated by a symbol; solid lines join the data points

**Figure 4** Normalized voltage $V/V_0$ versus time. The symbols corresponds to different injected currents (see legend) and take place one data point over 10. (inset) Relation between the relaxation time $\tau$ and the injected current
Figure 1: Dorbolo-JR03-1539

Figure 2: Dorbolo-JR03-1539
Figure 3: Dorbolo-JR03-1539

Figure 4: Dorbolo-JR03-1539