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Temporal evolution of electrical resistance through the granular packing of Ni beads

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Abstract: In this paper we investigate the temporal evolution of the electrical resistance through different two-dimensional (2D) packings of Ni beads when 1 mA current is injected in them. In the first stages of measurements, resistance decreases towards a saturation value and it can be fitted with Mittag-Leffler function. Fitting parameters show that the relaxation dynamics does not depend on the type of the packing. Different packings show differences in the initial values of the resistance which is attributed to the formation of new microcontacts during the formation of the new packing. Pauses in the flow of the current cause the resistance either to decrease, increase or remain the same, depending on the packing. Longer measurements show, that after the initial drop, the resistance starts to rise which can probably be attributed to the deterioration of microcontacts between the beads. Small variations in temperature do not affect the temporal evolution of the resistance.

INTRODUCTION

The electrical properties of metallic granular packings differ from the properties of the bulk samples of the same material. When an electric current is imposed on such a system, its voltage is not unambiguously determined by the current flow through the packing, i.e. the current-voltage dependence is not linear, but shows hysteresis properties and memory effects (Falcon & Castaing, 2005; Falcon & Castaing, 2005; Vandembroucq et al., 1997; Dorbolo et al., 2002; Falcon et al., 2004).
Previous research has identified several important factors causing this phenomenon: grain to grain contact is imperfect because of the surface roughness, electrical paths are created in the conducting states and geometrical variables such as the system dimensionality are to be taken into account (Dorbolo et al., 2002).
Tests of the change in electrical resistance over time have shown the resistance drop, which is initially more intense and later it slows down. Temporal evolution of resistance R(t) in metallic granular packings when a stable current through the system is established can be found only in a few papers (Yoon et a., 1999; Dorbolo et al., 2003; Lee et al., 2007; Jakšić et al. 2017). The decrease in resistance is considered to be the result of improved micro contacts (Toler et al., 2013) between the granules due to their local welding. Therefore, the electrical conductivity of granular packing is sensitive to mechanical and temperature perturbations (Vandewalle et nal., 2001; Bonamy et. al., 2000). Examining the temporal evolution of resistance in metallic granular packings of different materials can help to better understand the mechanisms behind it.

EXPERIMENTAL

Temporal evolution of electrical resistance R(t) was recorded for 2D granular packings of 52 Ni beads with radii 10.3 ± 0.02 mm. Surface roughness of the beads varied between 1.6-6.3 μm corresponding to N7-N9 roughness classes. Ni beads were placed in a rectangular box with dimensions 13.5 cm x 3.3 cm made out of plastic and wood. Brass electrodes were mounted along the lateral sides of the box, as presented in Figure 1.

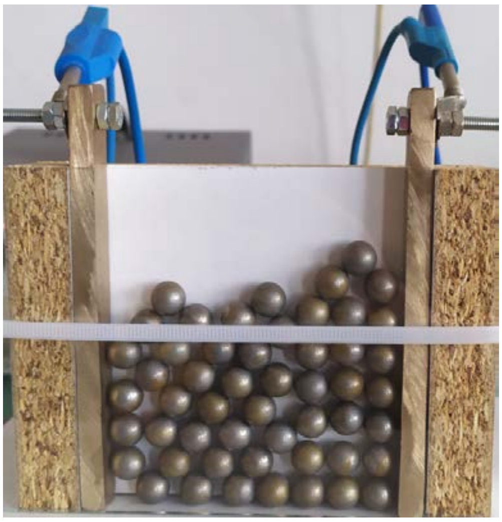


Figure 1. Experimental set-up for 2D packing of Ni beads.

The measurements were conducted for different 2D packings of the beads. The packing of the beads was established by the following procedure: the metallic grains were poured into the box which was initially placed in the horizontal position thus enabling all the beads to separate from each other (terminating contacts between all the beads). After that the box was straightened and then tilted under 8° inclination angle in respect to the horizontal position thus enabling us to visually control 2D ordering.

The box was placed on an anti-vibrational surface and the temperature in the room was measured using digital thermometer with recorded variations of 1.7 ℃ during measurements. The temporal evolution of the resistance was recorded by allowing a steady flow of current I = 1 mA from the current source Keithley 228 through the granular packing. The voltage was measured using Multimetar GDM 8621A (GW – Instek). The acquisition of data was carried out and controlled by a home-made computer program recording voltage with a 10 s step and calculating resistance using Ohms law. The resistance of each granular packing was measured for different time intervals.

Very strong contacts between all the components in the system were formed and any change in the voltage value recorded during measurements was caused exclusively because of the changes within the granular packing. This was checked by measuring the values of voltage between the electrodes and the measuring devices (the voltage was constant between the current source and the electrode and between the multimeter and the electrode).

RESULTS AND DISCUSSION

Resistance measurements were carried out on a number of different 2D packings of Ni beads through which a constant current I = 1 mA was injected. The measurements were performed right after the formation of a particular packing which lasted for a few seconds.

Figure 2 shows five representative measurements of the temporal evolution of the resistance for different packings during 1200 s. It can be concluded that the initial values of the resistance differ from each other regardless of the fact that we always used the same beads. This means that the initial resistance depends on the type of packing which can be attributed to creation of new contacts during the formation of a new packing (Jakšić et al., 2017). Also, the resistance decreases with time, faster in the first stages of relaxation and later on it slows down with a tendency of saturation.

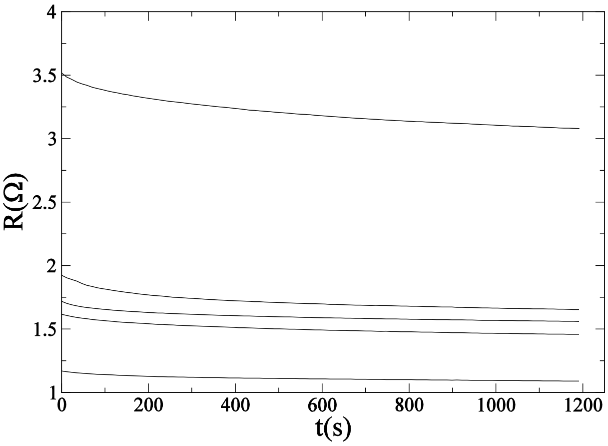


Figure 2: Temporal evolution of resistance for five different packings of Ni beads.

Our experimental data presented in Figure 2 can be fitted with Mittag-Leffler (ML) function of the form:

$$R(t) = R(\infty) - [R(\infty) - R_0]E_\alpha\left[-\left(\frac{t}{\tau}\right)^\alpha\right] \quad (1)$$

where $R(\infty)$ is asymptomatic value of resistance R(t) with $t \to \infty$, R_0 is the initial value of resistance, and E_α is ML function of order α, $0 < \alpha < 1$ (Haubold et al., 2011). ML function is defined through the inverse Laplace transform:

$$E_\alpha\left[-\left(\frac{t}{\tau}\right)^\alpha\right] = \lambda[(u + \tau^{-\alpha}u^{1-\alpha})^{-1}] \quad (2)$$

From which the series expansion can be obtained:

$$E_\alpha\left[-\left(\frac{t}{\tau}\right)^\alpha\right] = \sum_{n=0}^{\infty}\frac{(-(t/\tau)^\alpha)^n}{\Gamma(1+\alpha n)} \quad (3)$$

ML function interpolates between the initial stretched exponential form:

$$E_\alpha\left[-\left(\frac{t}{\tau}\right)^\alpha\right] \sim \phi_1 = \exp\left[-\frac{1}{\Gamma(1+\alpha)}(t/\tau)^\alpha\right], t \ll \tau \quad (4)$$

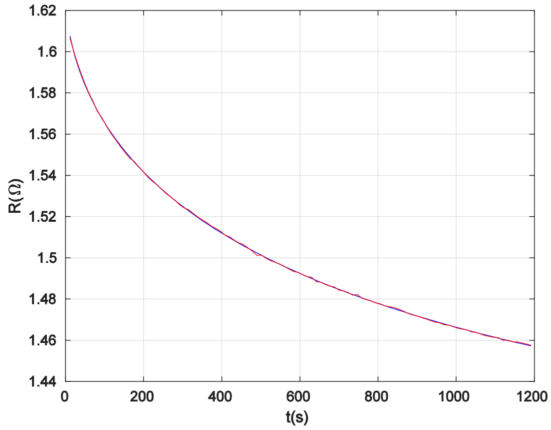
and the power behavior for later times:

$$E_\alpha\left[-\left(\frac{t}{\tau}\right)^\alpha\right] \sim \phi_2 = \frac{1}{\Gamma(1-\alpha)}(t/\tau)^{-\alpha}, \quad t \gg \tau \quad (5)$$

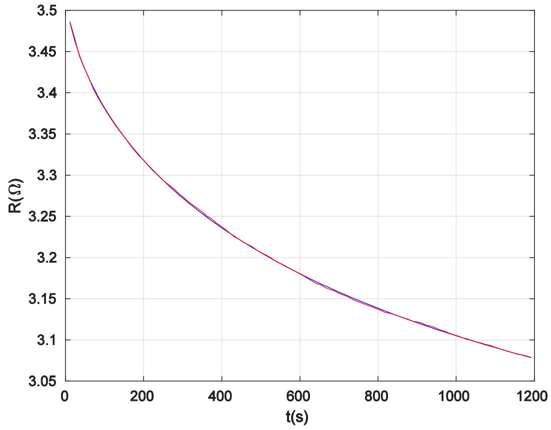
The same function was used in (Jakšić et al., 2017) for fitting the electrical conductivity for a constant flow of 1 mA current through a metallic cylinder packing.

Experimental results of R(t) (red line) fitted with ML function (equation (1), blue line) for three different packings of the beads, represented in Figure 3a-c show excellent agreement. The fitting parameters are as follows:

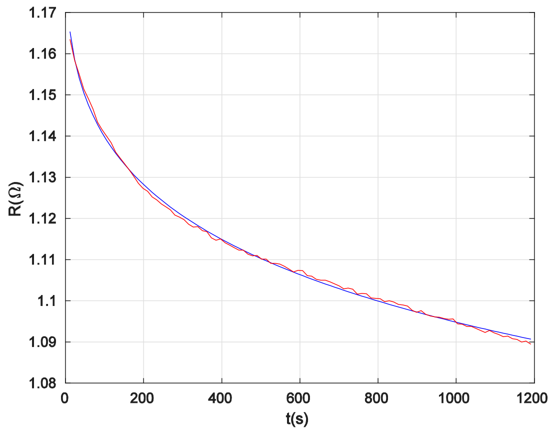
a) $\tau_1 = 1.5421 \cdot 10^3$s, $\alpha_1 = 0.57583$, b) $\tau_2 = 1.5410 \cdot 10^3$s, $\alpha_2 = 0.6255$ and c) $\tau_3 = 9.2724 \cdot 10^3$s, $\alpha_3 = 0.3535$. It can be concluded that the relaxation dynamics does not depend on the way in which the beads are arranged in the packing. For instance, two different packings can have the same relaxation dynamics ($\tau_1 \approx \tau_2$) and the evolution of the resistance toward the saturation value ($\alpha_1 \approx \alpha_2$), but on the other hand these processes can take place on a much different time scales ($\tau_3 \neq \tau_2$, $\alpha_3 \neq \alpha_2$) for different packings.



(a)



(b)



(c)

Figure 3: Temporal evolution of resistance for three different packings of Ni beads (red line), and Mittag-Leffler function fit of equation (1) (blue line).

In order to compare the rate of the lowering of the resistance for different packings, the time evolution of the normalized resistance R/R_0 (where R_0 is the initial resistance) was examined (Figure 4). It can be concluded that the change in the resistance can be the same for different packings but that is not a rule. Depending on the microcontacts that were established between the beads, the rate of the lowering of the resistance can either be faster or slower in different packings, which is in accordance with the results of M-L fitting.

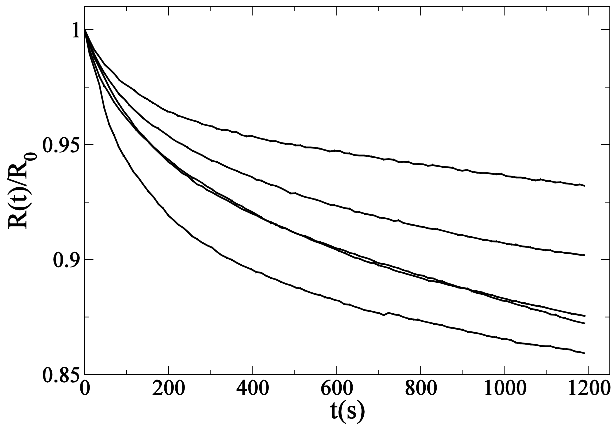


Figure 4: Normalized resistance versus time for different packings of the Ni beads.

Because every packing of the beads had different initial resistance, the measurements were conducted when the current was turned off for 10 s. These measurements were repeated for a number of different packings.

The typical results, presented in Figure 5, indicate that every break in the flow of the current can lead either to decreasing or increasing of the resistance, whereas in some cases the resistance remains unchanged. This means that in some cases the contacts between the beads were improved but in other cases they were weakened. The same breaks in the current flow were done consecutively on the same packing during longer measurements, which is presented in Figure 7a-d, where all three previously mentioned cases were also observed.

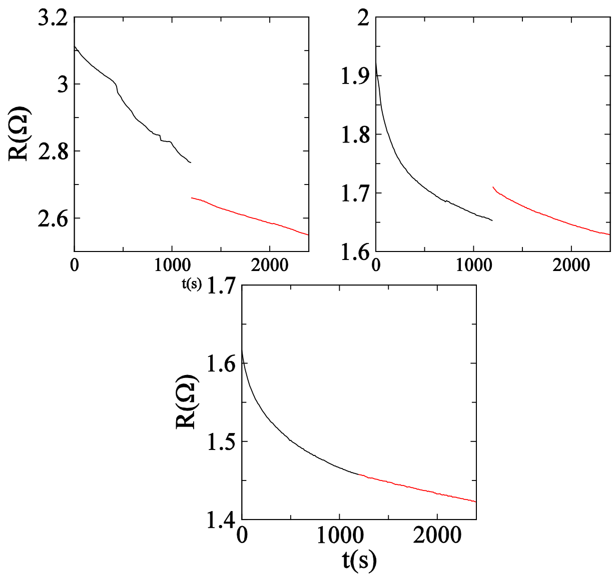


Figure 5: Temporal evolution of resistance during 2400 s, with the pause of 10 s after first 1200 s.

The same step changes in the resistance can be acquired if the current is turned off for longer periods of time. During these breaks, the conditions in the laboratory as well as the packing were not altered in any way. Figure 6 represents cases where the current was switched off for 30 minutes and 20 h respectively. The value of the resistance is either decreased or increased after the current is switched on again. The order of change in the value of the resistance is the same as in the case of the short breaks in the current flow.

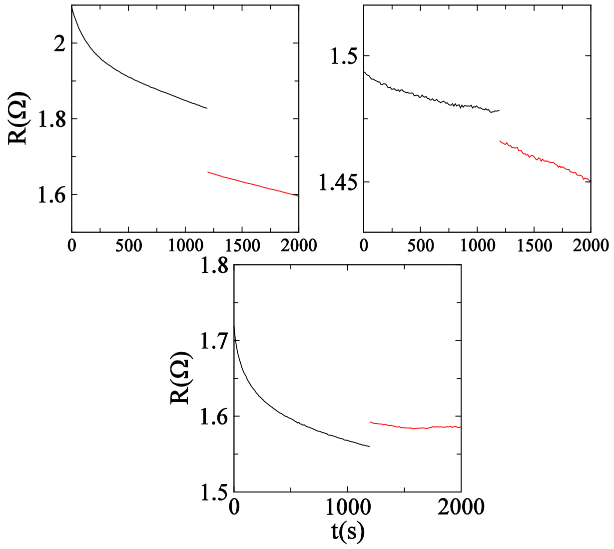
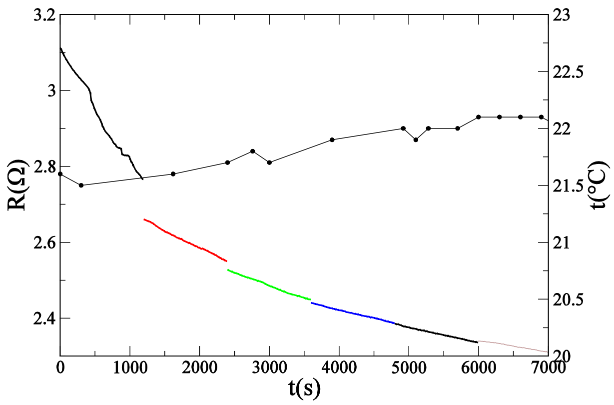
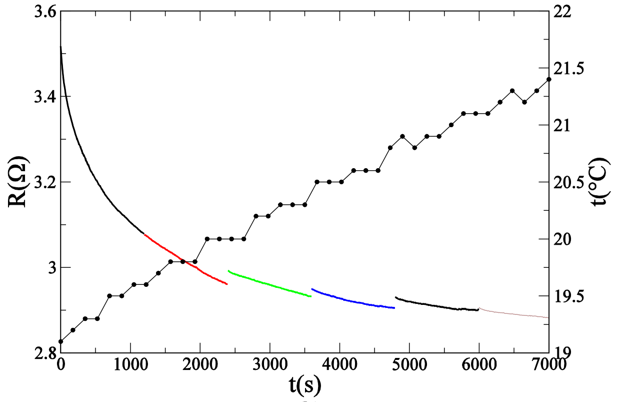


Figure 6: Temporal evolution of resistance during 2400 s, with 30 min pause (graph above) and 20 h pause (graph below) after the first 1200 s.

Figure 7a-d shows the typical resistance variations for four different packings. The measurements for the individual packing were performed during 7000 s, where after every 1200 s the current was switched off for 10 s. The slight temperature changes during the measurements were also recorded. Regardless of the fact that small temperature variations were recorded, resistance kept decreasing following every break in the current flow, which is in accordance with the results of the previous authors who kept the temperature constant throughout the measurements.



(a)



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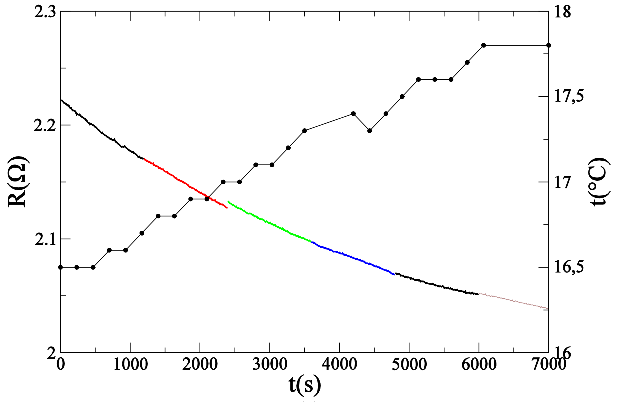

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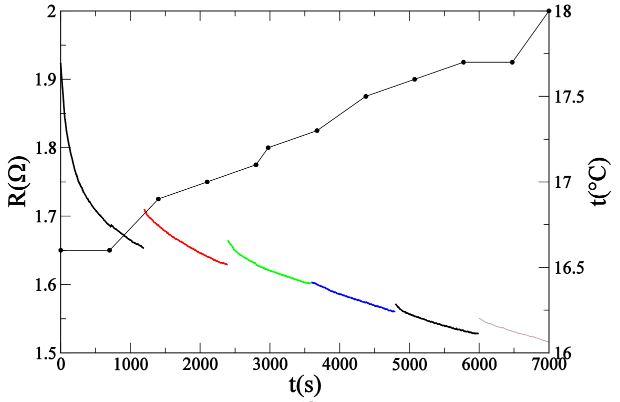

(d)

Figure 7: Temporal evolution of resistance with 10 s pauses after every 1200 s (left axis). Temperature variations during measurements (right axis).

A number of longer measurements that lasted up to 12000 s was also conducted and the typical results are presented in Figure 8. It can be seen that following the initial drop, the resistance starts to rise in the later phases of the experiments. We stipulate that this rise in resistance is caused because of the deterioration of contacts between the beads, but what causes this deterioration needs to be further examined.

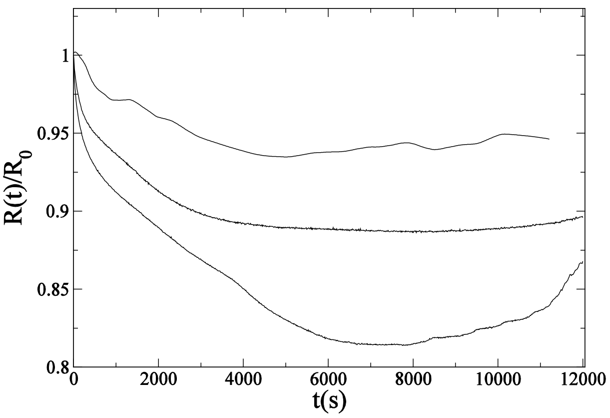

Figure 8: Temporal evolution of normalized resistance up to 12000 s long measurements

CONCLUSION

According to our results, the electrical resistance of a granular packing of Ni beads with 1 mA injected current drops in the first phases of relaxation towards a saturation value but later on it begins to rise which can be concluded from the longer measurements. This increase is not caused by external vibrations since it is well known that this kind of influence is recorded as an abrupt and step change in resistance during experiments. The decrease in the resistance in the first stages of measurements can be approximated with Mittag-Leffler function.
Pauses in the flow of the current through the packing can cause changes in the resistance, namely it can either increase or decrease.
Small increases of temperature do not affect the decrease of the resistance during relaxation.

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REFERENCES

Bonamy D., Laurent L., Claudin Ph., Bouchaud J.-Ph. and Daviaud F., (2000). Electrical conductance of a 2D packing of metallic beads under thermal perturbation, Europhys. Lett. 51, 614.

Dorbolo S., Ausloos M., and Vandewalle N., (2002). Hysteretic behavior in metallic granular matter, Appl. Phys. Lett. 81, 936.

Dorbolo S., Ausloos M., Vandewalle N., Houssa M., (2003). Aging process of electrical contacts in granular matter, J. Appl. Phys. 94, 7835.

Falcon E., Castaing B., Creyssels M., (2004). Nonlinear electrical conductivity in a 1D granular medium, Eur. Phys. J. B 38, 475–483.

Falcon E., Castaing B. (2005). Electrical conductivity in granular media and Branly's coherer: A simple experiment, American Journal of Physics 73, 302.

Falcon E., Castaing B. (2005). Electrical properties of granular matter: From "Branly effect" to intermittency, eBook Powders and Grains, ISBN 9780429107733

Haubold H. J., Mathai A. M., Saxena R. K., (2011). Mittag-Leffler Functions and Their Applications, Journal of Applied Mathematics, Article ID 298628, doi:10.1155/2011/298628

Jakšić Z. M., Cvetković M., Šćepanović J. R., Lončarević I., Budinski-Petković Lj. and Vrhovac S. B., (2017). The electrical conductance growth of a metallic granular packing, Eur. Phys. J. B 90: 108.

Lee J. J., Lee C. W., Yu I., Jung Y. K., Lee J., (2007). Slow relaxation of randomly packed hard spheres, J. Phys.: Condens. Matter 19, 356202.

Toler B. F., Coutu Jr 1 R. A., McBride J. W. (2013). A review of micro-contact physics for microelectromechanical systems (MEMS) metal contact switches, J. Micromech. Microeng. 23, 103001.

Vandembroucq D., Boccara A.C., and Roux S., (1997). Breakdown Patterns in Branly's Coheror. Journal de Physique III, EDP Sciences, 7 (2), 303-310.

Vandewalle N., Lenaerts C., Dorbolo S., (2001). Non-Gaussian electrical fluctuations in a quasi-2d packing of metallic beads, Europhys. Lett. 53, 197

Yoon S. S., Lee J. J., Yu I. (1999). Pattern formations in granular systems and their implications to dynamics of ferroelectrics, Journal of the Korean Physical Society, 35.

Summary/Sažetak

U ovom radu smo ispitali vremensku evoluciju električnog otpora u različitim dvodimenzionalnim (2D) pakovanjima Ni kuglica kada se kroz njih propusti struja jačine 1 mA. Naši rezultati pokazuju da u prvim fazama mjerenja otpor opada prema nekoj vrijednosti zasićenja. Eksperimentalni rezultati u prvim fazama mjerenja se veoma dobro mogu fitovati Mittag-Leffler funkcijom. Parametri fita pokazuju da dinamika relaksacije ne zavisi od tipa pakovanja tj. različita pakovanja mogu imati iste parameter fita. Pored toga, različita pakovanja pokazuju razlike u početnim vrijednostima otpora što se može pripisati formiranju novih mikrokontakata prilikom formiranja novog pakovanja. Duže ili kraće pauze u protoku struje uzrokuju promjenu otpora za određeno pakovanje. Naime u zavisnosti od tipa pakovanja kuglica, otpor može da poraste, opadne ili čak ostane nepromijenjen. Duža mjerenja pokazuju da nakon početnog pada, otpor počinje da raste, što se vjerovatno može pripisati pogoršanju mikrokontakata između kuglica. Male promjene temperature ne utiču na vremensku evoluciju otpora.