Slow Relaxation in Granular Compaction

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I Compaction Experiment

II Adsorption Theory

III Density Fluctuations

E. Ben-Naim, J. B. Knight, E. R. Nowak, H. M. Jaeger, S. R. Nagel, Physica D **123**, 380 (1998).

E. R. Nowak, E. Ben-Naim, J. B. Knight, E. Ben-Naim, H. M. Jaeger, S. R. Nagel, Phys. Rev. E **57**, 1971 (1998).

Compaction

- Uniform, simple system
- Slow density relaxation $\nu = 0$ Knight 95

$$\rho(t) = \rho_\infty - \frac{\rho_\infty - \rho_0}{1 + B\ln(t/\tau)}$$

- Parameters depend on Γ only

Disagreement with previous theories

- Void diffusion $\nu = 1$ Hong 93

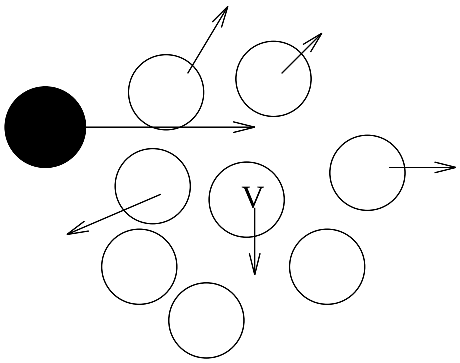
$$\rho(t) \cong \rho_\infty - At^{-\nu}$$

- Compactivity $\nu = \infty$ Mehta 93

$$\rho(t) \cong \rho_\infty - A_1 e^{-t/\tau_1} - A_2 e^{-t/\tau_2}$$

What causes logarithmic behavior?

Heuristic picture



$$\rho = \text{volume fraction}$$
$$V = \text{particle volume}$$
$$V_0 = \text{pore volume/particle}$$

$$\rho = \frac{V}{V + V_0} \qquad \text{or} \qquad V_0 = V\frac{1-\rho}{\rho}$$

Number of particles to be rearranged:

$$NV_0 = V \qquad \text{or} \qquad N = \frac{\rho}{1-\rho}$$

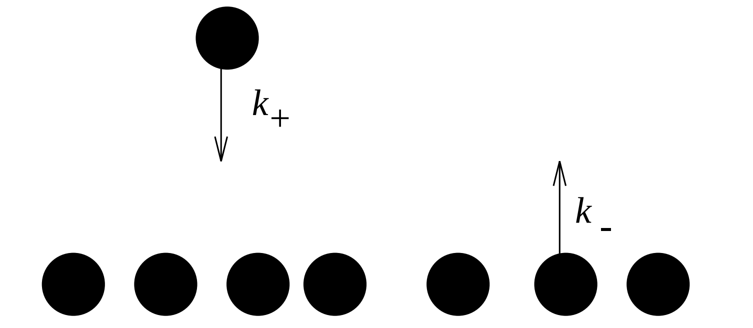
Exponential rearrangement time: $T = e^N = e^{\frac{\rho}{1-\rho}}$

$$\frac{d\rho}{dt} \propto (1-\rho)\frac{1}{T} = (1-\rho)e^{-\frac{\rho}{1-\rho}}$$

$$\rho(t) \cong 1 - \frac{1}{\ln t}$$

Volume exclusion causes slow relaxation

The "parking" model



- 1D Adsorption-desorption process
- Adsorption subject to volume constrains
- Desorption not restricted
- Detailed balance satisfied
- System reaches equilibrium steady state

Ignores: mechanical stability

Realistic: excluded volume interaction

Preliminary: lattice adsorption

Langmuir equation

$$\frac{\partial\rho}{\partial t} = -k_-\rho + k_+(1-\rho)$$

Steady state density ($\partial/\partial t \equiv 0$)

$$\rho_\infty = \frac{k_+}{k_+ + k_-} = \frac{k}{1+k}$$

Leading behavior ($k = k_+/k_-$)

$$\rho_\infty \cong \begin{cases} k & k \ll 1 \\ 1 - \frac{1}{k} & k \gg 1 \end{cases}$$

Relaxation ($\tau^{-1} = k_+ + k_-$)

$$\rho(t) = \rho_\infty + (\rho_0 - \rho_\infty)e^{-t/\tau}$$

No volume constrains, fast relaxation

Theory

$P(x,t) =$ Density of x-size voids at time t

$$1 = \int dx (x+1) P(x,t) \qquad \rho(t) = \int dx P(x,t)$$

Master equation:

$$\frac{\partial P(x)}{\partial t} = 2k_+ \int_{x+1} dy P(y) - 2k_- P(x)$$
$$+\theta(x{-}1)\left[\frac{k_-}{\rho(t)}\int_0^{x-1} dy P(y) P(x{-}1{-}y) - k_+ (x{-}1) P(x)\right]$$

Density rate equation:

$$\frac{\partial \rho(t)}{\partial t} = -k_- \rho(t) + k_+ \int_1 dx (x-1) P(x,t)$$

Convolution term assumes voids are uncorrelated (exact in equilibrium)

Equilibrium Properties

Exponential void distribution

$$P_\infty(x) = \frac{\rho_\infty^2}{1-\rho_\infty} \exp\left[-\frac{\rho_\infty}{1-\rho_\infty} x\right]$$

Mass balance

$$k_-\rho_\infty = k_+(1-\rho_\infty) \exp\left[-\frac{\rho_\infty}{1-\rho_\infty}\right]$$

Leading Behavior

$$\rho_\infty \cong \begin{cases} k & k \ll 1 \\ 1 - \frac{1}{\ln k} & k \gg 1 \end{cases}$$

0.95 coverage requires huge $k = 10^9$!

Volume exclusion dominates at high densities

The sticking probability

Total adsorption rate

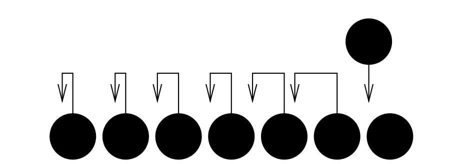
$$\int_1 dx(x-1)P_\infty(x) = k_+(1-\rho_\infty)\exp\left[-\frac{\rho_\infty}{1-\rho_\infty}\right]$$

Reduced adsorption rate $k_+ \to k_+ s(\rho)$

Sticking probability

$$s(\rho) = e^{-N} \qquad N = \frac{\rho}{1-\rho}$$

Heuristic picture is exact in 1D



Cooperative behavior in dense limit

Relaxation

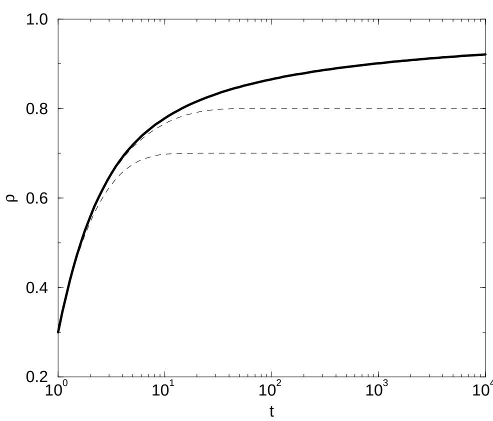
Quasistatic (near equilibrium) approximation

$$\frac{\partial \rho(t)}{\partial t} = -k_- \rho(t) + k_+ (1-\rho) \exp\left[-\frac{\rho}{1-\rho}\right]$$

I Desorption-limited case $(k_- \to 0)$

$$\rho(t) \cong 1 - \frac{1}{\ln k_+ t}$$

II Finite k_- $(\tau = [k_- \ln^2 k]^{-1})$



$$\rho(t) \cong \begin{cases} 1 - \frac{1}{\ln k_+ t} & t \ll \tau \\ \rho_\infty - A e^{-t/\tau} & t \gg \tau \end{cases}$$

Slow density relaxation

Examining the theory

- Diffusion similar to desorption
- Similar behavior in higher dimensions

Any predictive power?

$-$ Can not predict ρ_∞

$+$ Eventually - exponential relaxation

$+$ Test - Steady state density fluctuations

Distribution of equilibrium fluctuations

Multiple void distribution

$$G_\infty(x_1,\ldots,x_n) = \rho_\infty^{1-n} P_\infty(x_1) P_\infty(x_2)\cdots P_\infty(x_n)$$

For density ρ, $n=\rho L$, $V=\sum_i x_i = (1-\rho)L$

$$\begin{aligned} P_\infty(\rho) &= \int dx_1 \cdots \int dx_n G_\infty(x_1,\ldots,x_n)\delta\big(\textstyle\sum_i x_i - V\big) \\ &= \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left[-\frac{(\rho-\rho_\infty)^2}{2\sigma^2}\right] \end{aligned}$$

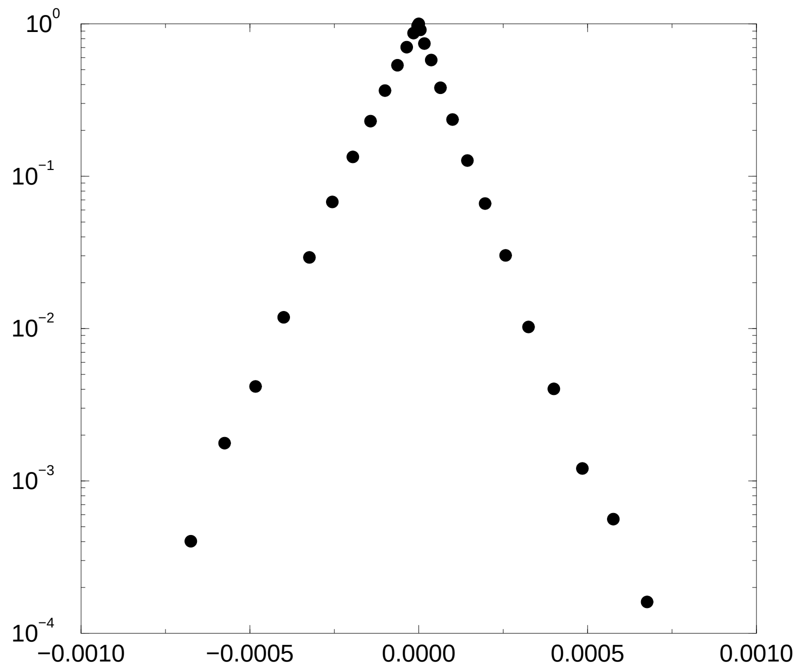
Variance

$$\sigma^2 = \rho_\infty(1-\rho_\infty)^2/L$$

Gaussian density fluctuations

Monte Carlo simulations

- Parameters: $k = 10^2$, $L = 10^3$.
- Theory: $\rho_\infty = 0.7719$, $\sigma^2 = 4.01 \times 10^{-5}$.
- Simulations: $\rho_\infty = 0.7718$, $\sigma^2 = 4.05 \times 10^{-5}$.



$P(\rho - \rho_\infty)$ versus $(\rho - \rho_\infty)^2 \mathrm{sgn}(\rho - \rho_\infty)$

$$\boxed{\text{Theoretical predictions verified numerically}}$$

Spectrum of density fluctuations

$$\mathsf{PSD}(f) = \left| \int d\tau e^{if\tau} \langle \rho(t)\rho(t+\tau) \rangle \right|^2$$

Leading behavior

$$\mathsf{PSD}(f) \cong \begin{cases} f^0 & f \ll f_L \\ f^{-\alpha} & f_L \ll f \ll f_H \\ f^{-2} & f_H \ll f \end{cases}$$

For noninteracting dilute case, linear theory, $\mathsf{PSD}(f) \propto [1 + (f/f_0)^2]$, with $f_0 = \tau^{-1} = k_+ + k_-$

In general, still open problem. Reasonable that $f_L = k_-$ and $f_H = k_+$

Similar noise spectrum for finite system Monte Carlo and experimental data

Conclusions

- Compaction dominated by exponentially rare grain size voids
- Growing time scales associated with cooperative bead rearrangements
- Argument is general - should hold for aspherical grains or horizontal tapping
- Gaussian density fluctuations

Outlook

- Glassy behavior
- Equilibrium hypotheses (Edwards)
- Compactivity - $\xi = (1-\rho)/\rho$ analog of T?
  $e^{-1/T}$ vs. $e^{-\rho/(1-\rho)}$ Edwards 89