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KINETIC MODEL OF COMPACTION IN GRANULAR MATERIALS

A simple kinetic equation describing the process of compaction relaxation to the asymptotic quasistationary state and satisfying the Carnahan–Starling equation of state has been formulated for the hard sphere model. In the framework of the Landau approach, we obtain the corresponding analytical solutions, which describe the homogeneous relaxation of the relevant order parameter in a sequential piecewise continuous series of intervals for the packing parameter. The characteristic relaxation time of the order parameter is found as a function of the model parameters. It is shown that the compaction can be satisfactorily described using the model of fractional kinetics, which reproduces the well-known asymptotic dependences in the corresponding limits. The results obtained agree well with the data of measurements concerning the compaction in granular materials under the action of external perturbations.

K e y w o r d s: granular materials, compaction, order parameter.

1. Introduction

Granular materials are conglomerates of a large number of microparticles repulsively interacting with one another. As a result, they lose some energy. Physical properties that characterize such systems under the influence of external perturbations create an appreciable domain of modern theoretical and experimental researches [1–4].

For instance, the compaction in granular materials under the action of external impacts, which has the efficient direct practical application, was studied in a number of works (see, e.g., works [5–12]) in detail. It was found experimentally that the kinetic phase diagram describing the compaction demonstrates an asymptotic saturation to quasiequilibrium states, which correspond to the maximum packing density or close to it. In the general case of the simplest systems (hard spheres), the relaxation law reproduces, with a sufficient accuracy, the known homogeneous Kolmogorov–Vogel–Fulcher scenario [6, 7]. Theoretical approaches to the packing kinetics – among them, it is enough to mention the kinetic model of free volume [7–9, 11, 12] – satisfactorily describe the asymptotic behavior of the corresponding order parameter in general. At the same time, a deficiency in approaches that would involve material relations between the physical properties and the structure peculiarities of examined systems impedes the parametrization of experimental data. In this work, we focus attention to the study of some relevant aspects.

As was shown in work [5], even the minimum specification in the description of the structure of systems with a complicated internal morphology (with granular materials belonging to this class) results in the formulation of an equation of state in the form of the Abel differential equation, which is not integrated by quadratures and allows solutions only in certain intervals of the values of packing parameter.

In this work, we consider a system of hard spheres satisfying the Carnahan–Starling equation of state [13], which is formulated in terms of compaction parameters. A simple kinetic equation for the correspondingly defined order parameter will be analyzed in the framework of the Landau approach [14, 15]. In the context of the proposed approach, the influence of parameters governing the state and the kinetics of the system – in particular, the compaction – on the characteristic times of the relaxation of the system to an asymptotic quasistationary state [8, 16] will be analyzed. The corresponding scenario of approaching the quasistationary state consists of piecewise continuous homogeneous relaxation cycles that cover the whole interval of the values of packing fraction.

2. Free Energy in the Carnahan–Starling Model of Hard Spheres

The Carnahan–Starling equation of state for hard spheres looks like [13]

$$PV = NkT\,\frac{1+\eta+\eta^2-\eta^3}{(1-\eta)^3}, \tag{1}$$

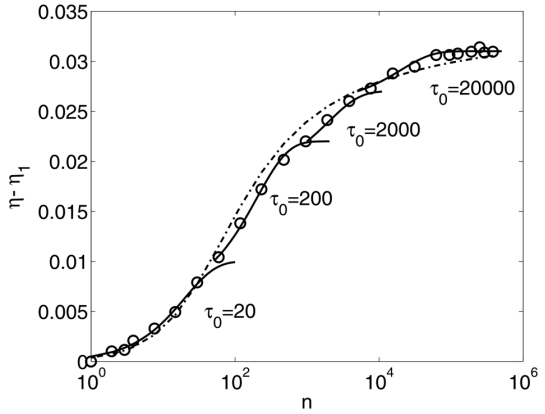
where P is the pressure, V the volume, N the number of particles, the product kT defines the particle energy scale, $\eta = \frac{1}{6}\pi\sigma^3\frac{N}{V}$ is the compaction parameter, and σ the particle diameter. Using the defining relation for the pressure, $P = -\left(\frac{\partial F}{\partial V}\right)_T$, the expression for the energy F in terms of the parameter η can be obtained from formula (1) in the following form:

$$F = NkT\left(\frac{3-2\eta}{(1-\eta)^2} - \frac{3-2\eta_1}{(1-\eta_1)^2} + \ln\frac{\eta}{\eta_1}\right). \tag{2}$$

Here, η is the current value of compaction parameter, and η_1 is the initial one. In the limiting case where the particle dimensions tend to zero, Eq. (2) results in the known relationship for the energy of ideal gas,

$$F = -NkT\ln\frac{V}{V_1}.$$

Experimental observations [7] show that the system asymptotically saturates (by packing) to a certain limiting quasistationary state (see Figure). In order



Experimental dependence (∘) of the packing $\eta-\eta_1$ in a granular system on the number of taps n [7], and the inverse-logarithmic (the dash-dotted curve) and piecewise continuous (the solid curves) approximations of experimental data making use of the simple exponential law of relaxation (12). The experimental data [7]: $\eta_1 = 0.56$ and $\eta_\infty = 0.591$

to study the process of relaxation near the saturated state, let us introduce the order parameter ϕ using the rule

$$\phi(t) = \frac{\eta_\infty - \eta(t)}{\eta_\infty - \eta_1}, \tag{3}$$

where $\phi(0) = 1$ corresponds to the initial state, and $\phi(\infty) = 0$ to the asymptotic quasistationary state of the system. This definition of order parameter allows us, in what follows, to apply the Landau approach [14], while describing the relaxation of the ordering field in a vicinity of the asymptotic stationary state.

The free energy F, which is given by relation (2) and does not involve fluctuation effects, can be rewritten as a function of the parameter η in the form

$$\frac{F(\phi)}{NkT} = \frac{3-2\eta_\infty+2(\eta_\infty-\eta_1)\phi}{(1-\eta_\infty+(\eta_\infty-\eta_1)\phi)^2} - \frac{3-2\eta_1}{(1-\eta_1)^2} +$$

$$+\ln\frac{\eta_\infty-(\eta_\infty-\eta_1)\phi}{\eta_1}. \tag{4}$$

Expanding $F(\phi)$ in a power series,

$$\frac{F(\phi)}{NkT} = A_0 + A\phi + B\phi^2 + C\phi^3 + D\phi^4 + ..., \tag{5}$$

and using Eqs. (4) and (5), we obtain the equation

$$\frac{F(\phi)}{NkT} = A_0 + \sum_{k=1}^{\infty}\phi^k(\eta_\infty-\eta_1)^k \times$$

$$\times\left[\frac{(3+k)-2\eta_\infty}{(1-\eta_\infty)^{k+2}}(-1)^k - \frac{1}{k\eta_\infty^k}\right], \tag{6}$$

where

$$A_0 = \ln\frac{\eta_\infty}{\eta_1} + \frac{3-2\eta_\infty}{(1-\eta_\infty)^2} - \frac{3-2\eta_1}{(1-\eta_1)^2}.$$

It can be shown that the sum in expression (6) is directly independent of the initial value of the hard sphere packing parameter η_1. The aforesaid testifies that the system can transform, by packing, from different initial states to the same asymptotic quasistationary one.

3. Kinetic Equation

Let us express the kinetic equation for the order parameter ϕ in the form

$$\frac{\partial\phi}{\partial t} = -\Gamma\frac{\delta F}{\delta\phi}, \tag{7}$$

where Γ is a kinetic coefficient. The analysis of the expression obtained for F demonstrates that the functions, which were obtained by confining series (6) to the quadratic terms inclusive, have one extreme point (namely, a minimum) in the physical region of the values of packing parameter η. In a vicinity of those states, we may expect a deceleration of the relaxation processes in the model system.

As an example, let us consider Eq. (7) with regard for series (6) confined to the quadratic contribution inclusive. The corresponding kinetic equation in the approximation described above reads

$$\frac{\partial \phi}{\partial \tau} = \alpha - \beta\phi, \tag{8}$$

where $\alpha = -A$, $\beta = 2B$, and $\tau = t\,(\Gamma NkT)$. Equation (8) has the trivial solution

$$\phi = \frac{\alpha}{\beta} + \left(1 - \frac{\alpha}{\beta}\right) e^{-\beta\tau}. \tag{9}$$

The substitution of Eq. (3) into formula (9) gives

$$\eta(\tau) = \eta_1 + (\eta_c - \eta_1)\left(1 - e^{-\tau/\tau_0}\right), \tag{10}$$

where $\tau_0 = \beta^{-1}$ is the characteristic relaxation time, and

$$\eta_c = \eta_\infty - (\eta_\infty - \eta_1)\frac{\alpha}{\beta}. \tag{11}$$

Note that, in the framework of the model concerned, the quantity η_c has to be determined self-consistently.

Equation (10) testifies that the relaxation process described by the simplified model constructed above satisfies the Kolmogorov–Vogel–Fulcher homogeneous scenario [7].

4. General Features of Compaction Parameter Relaxation

From the definition of the parameter τ_0 (see Eqs. (8)–(10)) in view of expressions (8), (5), and (6), it follows that $\tau_0 \backsim (\eta_\infty - \eta_1)^{-2}$. Hence, if the packing degree of the initial system increases ($\eta_1 \to \eta_\infty$), the characteristic relaxation time τ_0 grows and considerably slows down the packing process.

From relation (10), one can see that the packing parameter $\eta(\tau)$ asymptotically tends to the value $\eta_c \approx 0.647$. The deviation of η_c from $\eta_\infty = 0.74$ is associated with restrictions imposed on the function F, when the series expansion was confined to the square-law term. The account for terms in the F-expansion up to the 4-th order inclusive gives rise to a nonlinear equation. The solution of the latter in the first-order approximation, similarly to the case of the linear equation, demonstrates an asymptotic approach to the packing value $\eta_c \approx 0.62$. The account for terms up to the third-order one inclusive in Eq. (7) results in a differential kinetic equation with the square-law nonlinearity.

The nonlinear equations mentioned above can be solved in the linear approximation. The latter procedure gives rise to the appearance of corrections in the expressions for α, β, and η_c.

The results obtained demonstrate that, in the framework of the proposed model, the packing evolves following a nonmonotonic scenario. The applications of polynomials with even power exponents in the kinetic equation (7) testify that there are the certain values of packing parameter η_c, in whose vicinities the system comes closer to the asymptotic quasistationary state. At the same time, the application of odd-order polynomials in the kinetic equation (7) gives no solutions corresponding to the packing saturation at asymptotic states. Instead, they are well approximated by linear functions. This fact is associated with the presence of a minimum for even-order polynomials and its absence for odd-order ones in the physical (for the hard sphere packing) region. The positions of the minimum depending on the polynomial order are shown in Table.

The value $\eta_c = 0.647$, which was calculated by formula (11) with the use of the maximum possible packing value $\eta_m = 0.74$, falls within the limits of η_{RCP}-values (random close packing) from 0.63 to 0.65 known for the hard sphere model. Note that the Carnahan–Starling equation of state, which was used to obtain this result, does not make allowance for some factors actual in real granular systems, such as the energy dissipation, deviation of particles from the spherical form, state of their surface, size dispersion, intensity and type of a perturbation of the system,

Minimum position for various orders of polynomial $F(\eta)$

N	2	4	6	8	10	12	$\to \infty$
η_c	0.647	0.620	0.602	0.589	0.579	0.570	0.48

and presence of external fields (e.g., gravitation). In many experiments, the asymptotic values of packing in granular materials have different magnitudes [7, 17–22].

Therefore, in order to apply the obtained result (10) to the parametrization of experimental data, we may approximately change the calculated critical value of packing, η_c, by the asymptotic one, η_∞, which is observed, e.g., experimentally. Taking the remarks made above into account, a simple exponential law of relaxation can be given in the following form [1]:

$$\eta(\tau) = \eta_1 + (\eta_\infty - \eta_1)\left(1 - e^{-\tau/\tau_0}\right). \tag{12}$$

From Figure, one can see that the piecewise continuous approximation of experimental data by the exponential laws for the packing parameter η relaxation with characteristic times τ_0 that differ by almost an order of magnitude between the neighbor sections in the kinetic packing function allows the experiment to be satisfactorily described with the help of characteristic sections [23]. The latter result testifies to a nonmonotonic (nonlinear) character of the relaxation. Among the solutions of nonlinear kinetic equations that describe the relaxation of the packing parameter, we may specify, e.g., the so-called stretched exponential solution [7]

$$\eta(\tau) = \eta_1 + (\eta_\infty - \eta_1)\left(1 - e^{-[\tau/\tau_0]^\alpha}\right) \tag{13}$$

or the inverse logarithm [1]

$$\eta(\tau) = \eta_\infty - \frac{\eta_\infty - \eta_1}{1 + B\ln(1 + \tau/\tau_0)} \tag{14}$$

(see Figure). As a rule, those expressions are used for the parametrization of corresponding experiments. One of the adequate methods is the application of the fractional kinetic model to the description of the process of relaxation to the asymptotic quasistationary state. In the framework of this approach, the solution of the corresponding kinetic equation for the packing field looks like [22]

$$\eta = \eta_\infty - \Delta\eta E_\alpha\left(-\left(\frac{t}{\tau}\right)^\alpha\right), \tag{15}$$

where $\Delta\eta = \eta_\infty - \eta_1$, E_α is the Mittag–Leffler function of the order α ($0 < \alpha < 1$), η_∞ is the packing parameter in the asymptotic quasistationary state (in a vicinity of which, the free energy functional was expanded), η_1 is the packing parameter in the initial state, $\tau = k\Gamma$, Γ is the excitation parameter, and k the material constant. Asymptotically, as $\alpha \to 1$, the solution of Eq. (15) transforms into a simple exponential relaxation law (12):

$$\alpha \to 1; \Longrightarrow E_\alpha\left(-\left(\frac{t}{\tau}\right)^\alpha\right) \to \exp\left(-\frac{t}{\tau}\right), \tag{16}$$

which corresponds to the Landau–Ginzburg kinetic scenario of the relaxation in the linear approximation described above. In the case where the power exponent α tends to zero, the asymptotic of the Mittag–Leffler function transforms into the logarithmic one and describes, as was already remembered, the relaxation law for a slow compaction, which is observed in experiments on the packing in granular materials [1]. The results obtained testify that the packing and the mobility properties in granular materials are completely described in the framework of the fractional kinetic scenario. The process itself is inhomogeneous, by substantially depending on the interval of packing fraction values, in which it occurs.

5. Conclusions

The obtained results testify to a possibility of the formal application of the statistical physics methods to the description of certain phenomena in micromechanical many-particle systems (in particular, the compaction of granular materials, which are essentially dynamic dissipative systems) in a vicinity of their asymptotic quasistationary states. We also analyzed a possibility of using the model of fractional kinetics to describe the relaxation of the compaction parameter, which is more general in the case of the inhomogeneous density distribution.

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КІНЕТИЧНА МОДЕЛЬ
КОМПАКТИЗАЦІЇ ГРАНУЛЬОВАНИХ
МАТЕРІАЛІВ

Р е з ю м е

Для моделі твердих сфер, яка задовольняє рівняння стану Карнахана–Старлінга, сформульовано просте кінетичне рівняння, яке описує релаксацію фактора компактизації до асимптотичного квазістаціонарного стану. Користуючись підходом Ландау знайдені аналітичні розв'язки моделі, які описують однорідну релаксацію відповідно визначеного параметра впорядкування у послідовній кусково-безперервній низці інтервалів значень параметра впакування. Характерний час релаксації ступеня впорядкування визначено в термінах параметрів моделі. Показано, що задовільний опис компактизації може бути здійснений за допомогою моделі фракційної кінетики, яка у відповідних границях відтворює відомі асимптотичні закони релаксації. Отримані результати добре узгоджуються із даними експериментів, в яких безпосередньо вимірюється компактизація гранульованих матеріалів під впливом зовнішніх збурень.