COUPLED BURGERS EQUATIONS - A MODEL OF POLYDISPERSIVE SEDIMENTATION

SERGEI E. ESIPOV

James Franck Institute and Department of Physics, University of Chicago, 5640 South Ellis Avenue, Chicago, Illinois 60637, USA
ABSTRACT

This paper compares theory and experiment for the kinetics of time-dependent sedimentation. We discuss non-interacting suspensions and colloids which may exhibit behavior similar to the one-dimensional motion of compressible gas. The velocity of sedimentation (or creaming) depends upon the volume fraction of the constituting particles and leads to Burgers-like equations for concentration profiles. It is shown that even the bi-dispersive system of two coupled Burgers equations has rich dynamics. The study of polydispersive case reveals a continuous “renormalization” of the polydispersity. We compare the Burgers system evolution with the experimental results on mono- and polydispersive sedimentation. The influence of thermal fluctuations is briefly discussed.

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1. Introduction

The study of the motion of particles in a fluid goes back to Einstein and before that to Brown. Here we consider the effect of gravity upon the particles. If they are heavier than the surrounding fluid the resulting motion is called sedimentation; if lighter it is creaming. The conventional and simplest problem involves the evolution of initially uniform suspension or colloid. At the bottom (of the test tube) there appears the sediment, and at the top - water free of particles which is called supernatant. Between these regions there is the original suspension itself. Thus, two interfaces are created, and they spread and propagate towards each other. The description of the bottom interface invokes high volume ratios leading to “traffic”-like problems with possible jams and other instabilities whereby additional interfaces may emerge [1]. In this paper we address the evolution of the top interface which we can study by thinking about the dilute limit, in which the volume fraction of the particles is much less than 1. We apply the continuity equation which describes the conservation of species with concentration \( c(x, t) \) and current \( \mathbf{J}(x, t) \). Such an equation is well-known in the case of very small particles which experience Brownian motion. It can be taken from Landau & Lifshitz [2, §58,59]

\[
\partial_t c + \text{div}\mathbf{J} = 0 \tag{1.1}
\]

where the particle flux \( \mathbf{J} \) is produced by external forces (such as gravity), and also depends on gradients of concentration, pressure, and temperature. In the experiments which we discuss below, the influence of the (other than hydrostatic) pressure gradient and temperature gradient is negligible, so that the particle flux
can be written as an expansion in the concentration gradient in the form

$$J = \mathbf{V}(c) \cdot \nabla c - D(c) \nabla c,$$  \hspace{1cm} (1.2)

where the sedimentation velocity $\mathbf{V}(c)$ is produced by gravity and the hydrodynamic interactions, and the particle diffusivity $D(c)$ is produced both by thermal (Brownian) motion and hydrodynamic motion. Eqs.(1.1), (1.2) have not been applied quantitatively to sedimenting systems. It is instructive to discuss the possible reasons.

The kinetic coefficients $\mathbf{V}(c)$ and $D(c)$ are known for small enough particles when the Brownian motion dominates. These mixtures are called colloids. The velocity, $\mathbf{V}(c)$, has been calculated in the profound paper by Batchelor [3], see also [4]. Batchelor also found the dependence of Brownian diffusivity upon concentration [5]. Batchelor’s theory provides results for mono- and polydisperse suspensions. Yet, the task of plugging these results into Eqs.(1.1), (1.2) and comparing quantitatively its evolution with real experiments has not been performed. There have been some qualitative results, though. It was recognized [6,7] that Eqs(1.1), (1.2) with $c$-dependent velocity resemble very closely the famous Burgers equation of one-dimensional compressible flow [8,2]. This equation leads naturally to shock waves in which the non-linear velocity dependence on concentration is balanced by diffusivity. We suggest here that the shock waves are the mathematical realization of the observed interfaces. Some non-linear aspects of this effect were studied long ago by Kynch [9], who discussed the shock wave toppling in the absence of diffusivity, and more recently by Barker and Grimson [6], and by van Saarloos and Huse [7]. Baker and Grimson pointed out the fact that the Burgers equation has an analytical solution (if the concentration dependence of diffusivity is neglected) and
argued its qualitative applicability to a sedimentation experiment [10]. No quantitative comparison was done. The interesting paper by van Saarlos and Huse [7] used Burgers equation to discuss the sedimentation layers which are sometimes observed in the course of sedimentation. We shall discuss this phenomenon in a later paper [11].

Here we seek a comparison between the time-dependent evolution predicted by Burgers equation and real experimental data [12-14]. We shall also extend the description of sedimentation within the framework of Burgers equation to suspensions with large non-Brownian particles. Some unresolved theoretical issues arise in this area. For large enough particles the Brownian diffusivity is surpassed by the so-called hydrodynamic diffusivity. This effect has been studied by different experimental groups [12,14-17] but has not been fully understood theoretically, although a number of steps in this direction has been made [18-20]. Interesting numerical results have been reported by Ladd [21] for model system consisting of 32 and 108 particles. The value of the hydrodynamic diffusivity depends upon unknown density-density correlation function of a suspension. This correlation function is not short-ranged as in the case of small enough particles (colloids) - the property used in the calculations of Batchelor [3,4] - and the dependence $V(c)$ for suspensions is also unknown theoretically. We avoid the unresolved theoretical issues by making a simple order of magnitude estimate of hydrodynamic diffusivity which is different from previous estimates and consider the numerical prefactor as a fitting parameter. We also ignore the tensor-like structure of hydrodynamic diffusivity which is hopefully insignificant in the one-dimensional description offered by Eqs(1.1), (1.2) for concentration field depending solely upon vertical coordinate. As for the velocity of hindered settling, it will be described by an empirical dependence.
We would like to go further in applying the Burgers equation. This paper contains a straightforward generalization to the systems of coupled Burgers equations which are capable of describing realistic polydispersive suspensions. We find the steady-moving solutions of coupled Burgers equations and reproduce the evolution of the suspension interface discussed originally by Smith [23] and, in implicit form, by Davis and Acrivos [24]. The coupled Burgers equations predict an interesting phenomenon, which we named phase shifts, it may be already observed in a bidispersive system. Imagine that we study the evolution of initially homogeneous bidispersive system. In a while there will be two steady-moving interfaces, corresponding to the two given species. Their velocities can be found by using Smith formulas [23]. What is the distance between these interfaces? This distance cannot be found by multiplying the velocity difference by elapsed time. One has to account for the stage of “disentanglement” of the interfaces interacting via the non-linear coupled Burgers equations. Thus, the distance acquires the mentioned phase shift which is of order of combined diffusive length calculated at the disentanglement time, i.e. it is a diffusivity-dependent effect. This effect is important for calculation of the width of interfaces of polydispersive suspensions. On the other hand the quantitative definition of phase shifts is not really straightforward: this phenomenon becomes important when an experimentalist can spatially resolve the interface profile, and possible definitions contain a fair degree of uncertainty. For example, one may expect a definition of the distance between the interfaces as a measure between the points where the concentrations of corresponding species diminish in, say, $e$ times. However, in the course of sedimentation in real polydispersive systems the segregation of particles improves gradually with time, and it is unclear which collection of species is to be traced with the above definition. We then
regard the solution of the coupled Burgers equations as a necessary step in describing the spreading of the interface. In the case of real polydispersive suspensions the distribution function of particle sizes $a, c(a, x, t)$ replaces the concentration. This distribution function evolves in a complex way near the interface. Step by step the system eliminates particles with different $a$. We call this evolution renormalization since it is produced by applying many times a given rule of eliminating the fastest species (infinitely many times in the continuum limit).

We introduce the Burgers equation in Section 2 and discuss the relevant kinetic coefficients. To show how this equation works, the experimental data of Ref.13 for a colloid are compared with the analytical solution. In Section 3 we used coupled Burgers equations to describe polydispersive systems. We first address the unique steady-state motion in terms of simple analytical formulas borrowed from physics of one-dimensional compressible gas [2] and then simulate the bi-dispersive case numerically to investigate the evolution of the initial condition problem. Description of the continuous polydisperse case is presented in Section 4. In the polydispersive case sedimentation leads to the continuous renormalization of the particle size distribution function as one scans through the top interface. The late-time solution is discussed together with the effects of diffusivity. Comparison with experiments [12,14] is given. Section 5 contains brief discussion of influence of intrinsic thermal fluctuations on the sedimenting system and indicates the connection with dynamical critical phenomena.
2. Monodispersive sedimentation

Depending on the particle size one distinguishes suspensions and colloids for a given fluid. The boundary is defined somewhat arbitrarily through a Péclet number,

\[ Pe = \frac{2aV_0}{D_0} = \frac{8\pi g \Delta \rho a^4}{3k_B T}, \]

which describes competition between action of (say) gravity and thermal fluctuations on a spherical particle of radius \( a \). Gravity results in particle downward motion with the Stokes velocity \( V_0 \),

\[ V_0 = \frac{(4/3)\pi a^3 g \Delta \rho}{6\pi \eta a} = \frac{2a^2 g \Delta \rho}{9\eta}, \]

due to the density difference \( \Delta \rho \) in a fluid with with viscosity \( \eta \) in the presence of gravity acceleration \( g \). Thermal fluctuations are the source of the Brownian diffusivity,

\[ D_0 = \frac{k_B T}{6\pi \eta a}, \]

where \( k_B T \) is the temperature in energy units. Systems with \( Pe < 1 \) are conventionally called colloids. Thermal fluctuations are strong for colloids and lead to a uniform density-density correlation function which is short-ranged. In the opposite case the structure and kinetic properties are exclusively determined by the interparticle hydrodynamic interactions. For water at room temperature and particles less that \( 1\mu m \) Brownian effects usually dominate, while larger particles form suspensions.

Evolution of the interface which separates the region of the initial distribution and supernatant in colloids can be described by the concentration profile \( c(x, t) \).
which obeys a Burgers-like equation (see above)

\[
\partial_t c = \partial_x [V(c)c] + \partial_x [D(c)\partial_x c],
\]

(2.4)

where \(V(c), D(c)\) represent the Stokes velocity and gradient diffusivity modified by the presence of other particles. The axis \(x\) is directed upward, opposite to the direction of gravity. If \(c(x, t)\) is normalized to be the volume fraction \((c = \frac{4}{3}\pi a^3 n\) with \(n\) being the number density), then

\[
V(c) = V_0 f_v(c),
\]

(2.5)

where \(f_v(c)\) is the hindering effect and

\[
f_v(c) = 1 - kc + O(c^2), \quad c \ll 1.
\]

(2.6)

According to calculations by Batchelor [3], \(k \approx 6.55\) in the dilute limit, \(c \ll 1\). Larger values of the concentration are sometimes approximated, for example, by the Richardson-Zaki empirical formula [22], \(f_v(c) = (1 - c/c_0)^{k_{c0}}\). (There exist other suggested formulas [13].) Here \(c_0 < 1\) is the volume fraction at dense packing.

The diffusivity \(D\) entering (2.4) can be written as

\[
D_b(c) = D_0 f_b(c),
\]

(2.7)

with (again according to Batchelor [5], see also [25])

\[
f_b(c) = 1 - (k - 8)c + ... \approx 1 + 1.45c, \quad c \ll 1.
\]

(2.8)

The nonlinear terms in the expansion (2.8) (studied experimentally [25]) lead to a maximum of \(f_b\) at \(c \sim 0.15\), so that the entire concentration dependence of the diffusivity is within 10% of its bare values up to the volume fractions \(c \sim 0.3\).
Addressing now suspensions, we recall that the right-hand side of Eq(2.4) represents a double expansion of the particle current in terms of small concentration value and small spatial gradients of concentration. In the case of large Péclet numbers, we deal with strongly interacting systems, and the applicability of Eq(2.4) is unclear. To be more specific we do not doubt that the term $V(c)c$ must enter the particle flux $J$ in the homogenous suspension, see Eq(1.2), however the existence of a local (and/or fickian) expression for the inhomogeneity-induced part of flux has not been established. In fact, the numerical simulations by Ladd [21] indicate that motion of a marked particle is somewhat non-gaussian. Nevertheless, in this paper we use Eqs(1.2) and (2.4) to model the evolution of suspensions. Then, the function $D(c)$ is conventionally called hydrodynamic diffusivity [19]. This concept is an attempt to get the main effect of multiparticle forces without a very detailed calculation and can only be justified a posteriori, as it is sometimes the case with the Fokker-Planck method.

Thus, the hydrodynamic diffusivity of suspensions, which will be denoted as $D_h(c)$, reflects the net effect of the velocity fluctuations due to interparticle hydrodynamic interactions. This problem is reminiscent of those which arise in the description of dense plasma [20]. However, hydrodynamic forces decay with particle-particle separation like $1/r$ and provide even stronger interaction as compared to $1/r^2$ Coulomb forces. For Coulomb forces it is known (§27,41 in Ref.26) that plasma experiences collective and chaotic motions, the latter due to interparticle collisions. Hydrodynamic collective modes are dissipative, so the interesting question is the account for the collisions. Collisions in 3D plasma that are of most importance correspond to the small angle scattering events (§41, Ref.26, see also [27]). In the course of these events particles approach each other at a distance of order of
the Debye screening length although the interparticle separation is much smaller than that. Mean “free” path is inversely proportional to the volume fraction $c$, $l \sim 1/na^2 \sim a/c$. Diffusivity is the product of the mean velocity fluctuations, $v$ and $l$, $D(c) \sim av/c$. This is the $c$-dependence which is valid for usual gases (§7,8, Ref.26), interacting plasma (§43, Ref.26), and it was claimed by Koch and Shaqfeh [20] for hydrodynamic diffusion in the dilute limit.

Unfortunately, the pair collision approximation, which was developed by Landau for plasma (§41, Ref.26) does not work here: for the particles with equal densities and of identical size there is a well-known reversibility and symmetry (see, for example, the concise discussion by Hinch [19]). Namely, the solution for viscous flow surrounding two descending spheres provides no hydrodynamical repulsion and/or attraction, and the inter-particle separation remains constant in time. Thus, at least triple collisions are needed, and a question immediately arises whether it is legitimate to use triple collisions and disregard high order ones? Analogous situations do arise in plasma [28].

Koch and Shaqfeh argued that a particle distribution without correlations is a solution of the Liouville continuity equations describing all particles in a point-particle approximation. They continued that finite particle sizes are important to destroy this (say, initial) uniform distribution. Then Koch and Shaqfeh concluded that the most important triple collisions are those when two out of three particles are within $O(a)$ distance while the third particle is far away. We do not see any reason for such a conclusion. This conclusion also implies that there exists a way to resolve the question of higher order collisions without identifying a small parameter different from $c \ll 1$. In plasma physics this additional parameter is the condition of high temperature, $T \gg e^2/\bar{r}$ (§27, Ref.26). Namely, the plasma temperature
must greatly exceed the Coulomb interaction at the interparticle separation $\bar{r}$. This condition ensures that the Debye radius greatly exceeds $\bar{r}$, justifies the Landau collision integral and so on. At present we do not think it is possible to extend these ideas to the physics of sedimentation. Interestingly, the numerical results by Ladd [21] do indicate some screening, although it is quite different from what is presented in [20].

Hinch [19] pointed an argument by Caflish that uniform distribution with the finite particle sizes leads to convective flows due to density fluctuations. Hinch speculated that the convection will cease at the level of mixing which is comparable to the interparticle separation, $ac^{-1/3}$. The velocity fluctuations of the “remaining” convection are then of the order of the main term of the interparticle force, $V_0c^{1/3}$. We may continue this speculation by multiplying these two values,

$$D_h(c) = k'aV_0, \quad c \ll 1. \quad (2.9)$$

where $k'$ is the pure number. One may expect that in the zero concentration limit the effect of hydrodynamic diffusivity vanishes. This does not necessarily imply that $D_h(c)$ must go to zero, since the required system size and time needed to establish the steady-state regime both diverge as the concentration goes to zero. Using (2.9) one finds that the ratio $D_h/D_b$ is of the order of the Péclet number. If the dependence on concentration in Eq(2.9) is incorrect, there would exist an additional class of mixtures which cannot be ascribed to either suspensions or colloids.

Experimentalists have been analyzing hydrodynamic diffusion by two different techniques. One approach [12,14] is to investigate the gradient diffusion. The other is to trace a marked particle and evaluate the self-diffusivity [15-17]. In a
strongly interacting system the two approaches may, in principle, produce different values. Ham and Homsy [15] found that for the case of self-diffusion the factor $k'$ depends upon concentration for small $c < 0.01$, and saturates at about $k' = 5$ for larger values of $c$. Experiments by Nicolai et al discriminate between vertical and horizontal self-diffusivities and indicate $k' = 6 - 10$ for the former. There is a possibility that the whole tensor may be inferred from experimental data by Nicolai et al. As for the gradient diffusion analysis it will be discussed in detail below. We shall estimate $k' = 10$ from analyzing gradient diffusion of the experiments [12,14] (see Section 4). This estimate is within the error bars of the experimental estimates of self-diffusivity. The $c$-dependence of $D_h$ was not really tested in this paper since both the degree of polydispersity and uncertainty of the size distribution function used in [12,14] are found to be too large for this purpose.

We have introduced the Burgers equation and corresponding kinetic coefficients and, before we begin to use it, let us recall two approximations which are usually made when deriving equations (2.4)-(2.8).

1. The particles are supposed not to interact in any fashion apart for the hydrodynamic interaction and excluded volume effect, i.e. these particles are hard spheres. Such an approximation rules out many interesting systems exhibiting spinodal decomposition and phase interface formation [29].

2. Thermal fluctuations are neglected in Eq(2.4) except for the mean-field contribution to diffusivity $D_h$. These are “three-dimensional” fluctuations which have to be averaged over the lateral plane for each height $x$ of the sedimenting system. Alternatively, one can consider the 3D-version of Eq(2.4) with anisotropic drift in the vertical direction. As discussed by van Saarloss and Huse [7], the conservative noise leads only to small corrections (see also Section 5).
Now we attempt a detailed comparison with the experiment. It is well-known \cite{8} that for any smooth initial distribution $c(x,t)$ with the boundary conditions $c(-\infty,t) = c_0$, $c(\infty,t) = 0$ the late-time asymptotic is a single Burgers shock moving with a constant speed and width. Indeed, substituting $c(x,t) = c(x+vt)$ into (2.4) and integrating from $x$ to $\infty$ one obtains

$$D(c) \frac{dc}{dx} = [v - V(c)]c, \quad (2.10)$$

which for large negative $x$ gives $v = V(c_0)$. The shape of the interface can be found by another integration of Eq(2.10). This shape depends upon Péclet number. Using (2.5)-(2.8) we find an implicit equation for the concentration dependence on $x$

$$\left(\frac{8}{k} - 1\right) \log(c_0 - c) - \frac{1}{k c_0} \log \left(\frac{c_0 - c}{c}\right) = \frac{x P e}{2a}. \quad (2.11)$$

At intermediate times the experimentally resolved interface profile is time-dependent. We found relevant measurements in a paper by Al-Naafa and Selim \cite{13} who studied the evolution of the optical density of a monodisperse colloid. To make the comparison, we solve Eq(2.4) with initial condition $c(x,0) = c_0 \theta(x)$ to get the evolution of concentration

$$c(x,t) = \frac{c_0 f_1(x,t)}{f_2(x,t) + f_1(x,t)},$$

$$f_1(x,t) = \exp \left\{ - \frac{V_0 k c_0}{D_0} [x + V_0 t (1 - k c_0)] \right\} \left\{ 1 - \text{erf} \left[ \frac{x + V_0 t (1 - 2 k c_0)}{2 \sqrt{D_0 t}} \right] \right\},$$

$$f_2(x,t) = 1 + \text{erf} \left[ \frac{x + V_0 t}{2 \sqrt{D_0 t}} \right]. \quad (2.12)$$

We then try to fit the data in Fig.5, paper \cite{13} using this formula. The result is shown in Fig.1. The experimental parameters are given in the figure caption.
Note that there are no adjustable parameters used. Initially the shape of the optical density profile is more narrow than the prediction of formula (2.12). After a transient we get the optical shape which does fit.

We do not have any explanation of the transient. Our attempt to use more complex fit (see Section 4 for the description of polydispersive fit) by taking into account the small polydispersity of the suspension (±4.5 nm) did not result in significant changes. Thus the experiment both supports and denies the validity of Burgers equation for sedimentation. We need more experimental data to help resolve this issue.

3. Bidispersive sedimentation

We then consider the case of bimodal distribution of particle sizes. Let $c_1$ and $c_2$ be the concentrations of these particles, both obeying continuity equations (1.1). The corresponding fluxes $J_1, J_2$ are

$$J_i = V_i(c_1, c_2) - D_i(c_1, c_2) \frac{\partial c_i}{\partial x}.$$  \hspace{1cm} (3.1)

Using expansion at small concentrations we arrive to the two coupled Burgers equations

$$\partial_t c_1 = V_1 \partial_x [(1 - k_{11} c_1 - k_{12} c_2) c_1] + D_1 \partial_x^2 c_1$$

$$\partial_t c_2 = V_2 \partial_x [(1 - k_{21} c_1 - k_{22} c_2) c_2] + D_2 \partial_x^2 c_2.$$  \hspace{1cm} (3.2)

This system is a straightforward generalization of the monodispersive Burgers equation (2.4). The hindering velocity of each of the particle species depends linearly on both volume fractions. This dependence has been studied by Batchelor and Wen,
see Ref. 4 where one can find method of computing the constants $k_{11}, k_{12}, k_{21}, k_{22}$.

The dependence of diffusivities $D_1, D_2$ upon particle concentrations will be ignored in this Section together with the cross-terms when a gradient of, say, $c_1$ influences a flux of $c_2$ particles. These refinements (all vanishing with $c$) can wait until the validity of Burgers equation is understood.

First, we are looking for steady-moving solutions $c_{1,2}(x - v_{1,2}t)$. Substituting these into equations (3.2) and integrating once we have

$$D_1 \frac{dc_1}{dx} = (v_1 - V_1)c_1 + V_1 k_{11} c_1^2 - V_1 k_{12} c_1 c_2$$

(3.3)

and an analogous second equation. Two shocks may be observed for the relevant boundary conditions $c_{1,2}(-\infty) = c_{01,02}, c_{1,2}(\infty) = 0$. We then have to decide which of the waves is faster. Without loss of generality let us assume that $v_1 > v_2$ where

$$v_1 = V_1(1 - k_{11}c_{01} + k_{12}c_{02}),$$

(3.4)

$$v_2 = V_2(1 - k_{21}c_{01} + k_{22}c_{02}),$$

(3.5)

This leads to the first shock velocity to be $v_1$. The substance 2 changes its concentration when the first shock passes through. Let us denote this changed (we shall also call it renormalized) concentration as $c^*_2$. At the top of substance 1 we essentially have the problem of monodispersive sedimentation considered in previous Section. Let the hindered velocity of settling be $v^*_2$, as we have seen above, it is also the velocity of the second shock. Concentration $c^*_2$ obeys the quadratic continuity equation in the reference frame moving with the first shock velocity $v_1$.
(see Ref.2, Chapter IX, and Ref.23)

\[ c_2(v_2 - v_1) = c_2^*(v_2^* - v_1), \]  
\[ (3.6) \]

Here \( v_2^* \) is

\[ v_2^* = V_2(1 - k_{22}c_2^*). \]  
\[ (3.7) \]

Fig.2 illustrates the steady-state geometry of the system. It can be easily shown that if \( v_1 > v_2 \) then \( v_1 > v_2^* \), and \( v_1 \to v_2^* \) when \( v_1 \to v_2 \). Consequently, no other steady-moving solutions exist for this system. Thus, the steady-moving solution of coupled Burgers system are characterized by the well-known formulas for the interface (shock) velocities [23,25,30]. It is nice to get to know that these formulas also describe the solutions of coupled Burgers equations. Experimental confirmation of shock velocities predicted by Eqs(3.6), (3.7) for bidispersive (and tridispersive) systems can be found in Refs.25,30.

The coupled Burgers equations also allow to study the transient regimes prior to the establishing of the steady-moving shocks. Transient regimes were not reported experimentally, most probably due to the absence of relevant theoretical method of analysis, and we have no available experimental data to fit the concentration profiles. We hope to encourage studies of time-dependent phenomena by addressing physics of coupled Burgers equations. To give an example of transient behavior we solved the system of two coupled Burgers equations numerically.

The two shocks when formed separate linearly in time and their asymptotical shapes may be found by numerical integration of Eq(3.3) with real concentration dependences of the diffusivities, see [5]. Both shocks have finite widths (analytical formulas may be obtained in a number of limits) and quickly cease to interact,
see Fig. 2. It must be emphasized that no matter how simple the steady-moving solution of the system (3.2) may seem, the transition to this solution in time may be rich, see Fig. 3 where a change in coupling constant $k_{21}$ from 0.35 to 0.4 resulted in system inability to reach the dynamical steady-state within the integration time. Figs 2 and 3 show examples of the time dependent solutions of Eqs (3.2) which develop transient raises of concentration and even additional transient shocks (Fig. 3). The final profiles in Fig. 2 display the two regions discussed above: first shock at $x \approx 30$, accompanying with renormalization of $c_2$ from 1 up to 1.86; and the second shock at $x \approx 45$ at the end of the integration time. Measuring rate of change of the separation between adjacent profiles one can verify that the shock velocity is nicely predicted by Eqs (3.4), (3.7). However, the separation between the shocks accounts for the initial condition and interaction at the stage of shock “disentanglement” as well as for the widths of shocks.

It is also noteworthy that the described behavior is just an example of dynamics of a compressible one-dimensional gases.

### 4. Polydisperse sedimentation

It is not hard to apply the ideas of previous Section to the real polydisperse case. We shall first discuss the situation where finite number $N$ of particle sizes is involved and particle groups are numbered, $1 \leq i \leq N$. As before one should calculate the hindered velocities of sedimentation of all particle groups which are given by

$$v_i = V_i \left[ 1 - \sum_{j=1}^{N} k_{ij}c_j \right],$$

and represent a generalization of expressions like (3.4). Again we refer to Ref. 14.
for the constants $k_{ij}$. The maximal velocity $\max_i(v_i)$ is to be found and the corresponding substance (say, $i_0$) will form the lowest (fastest) shock. Generally speaking, the motion of the edge of the size distribution is not necessarily the fastest, one may rather say that some substance $i_0$ will be move down first, depending upon the matrix $k_{i,j}$ and size (and density) distribution function. Other particle concentrations at the top of the leader will be renormalized, according to the equations

$$v_i^* = V_i \left[ 1 - \sum_{j=1}^{j=N} k_{ij} c_j^* \right], \quad (4.2)$$

$$c_i(v_i - v_{i_0}) = c_i^*(v_i^* - v_{i_0}), \quad (4.3)$$

which form a system of quadratic equations. Note, that the system can be formally extended to $i, j = i_0$ given that $c_{i_0}^* = 0$.

Now the entire procedure is to be repeated $N$ times. At each level of renormalization one additional substance is eliminated and others get renormalized. An example can be seen in Fig.4 for the evolution of initially Gaussian-like size distribution function with $N = 26$ different particle sizes, and real parameters taken from paper [12]. In this particular case there is no inversion of ordering, i.e. larger sizes are eliminated first. The result of the calculation is a sequence of successively eliminated particle species and corresponding velocities. The interface shape becomes wider in time, although not as wide as one would obtain without renormalization. The concentration profile in the limit of vanishing diffusivity can be found by adding regions with renormalized concentrations at separations prescribed by successive velocity differences. Renormalization leads to thinner interfaces, this is the so-called phenomenon of self-sharpening of the interface [12,14].
when smaller particles at the top of larger particles move faster then they would do if mixed with larger particles. When diffusivity is finite - a contribution to the interface width comes from the phase shifts.

Before moving on to the continuous case we discuss the influence of diffusivity from yet a different angle. For a single Burgers shock connecting concentration change from $c_{-\infty}$ to $c_{+\infty}$ the width is given by

$$W = \frac{2D}{V_0 k(c_{-\infty} - c_{+\infty})}. \quad (4.4)$$

where for a moment we assume the simplest possible situation: constant diffusivity and monodispersive case. The smaller is the concentration difference the weaker is the non-linear effect competing with constant diffusivity [8], and the shock width becomes proportionally larger. Similar phenomenon exists in the polydisperse case, the larger is the number of different particle sizes which are taken into consideration - the smaller are the concentration changes, the smaller are the velocity differences. In addition, the widths of the successive shocks overlap. Thus, diffusivity provides a time-dependent cut-off for the achieved resolution in particle size step.

This physics is reflected in the renormalization of size distribution function. When particles of a given size move down and are not present at some height, the distribution function of this size at that height becomes zero (rigorously speaking it is, of course, of order of exponentially small, $exp(-Vx/D)$, diffusive corrections). In the limit of large $N$ this leads to a discontinuity which moves monotonously or jumps depending on absence or presence of ordering inversion discussed above. Discontinuous renormalization of the size distribution function implies that diffusivity is to be taken into account, and the existence of the diffusive time-dependent cut-off
regularizes the problem. At any point along the interface new structure appears as time goes on. The minimal resolved size step diminishes with time, and the emerging of the fine structure of the distribution function (or segregation) continues. Note that certain integrated properties such as the profile of the optical density may be still perfectly smooth under these circumstances, it is the size distribution function which changes most drastically within the suspension interface.

In the continuous case the coupled Burgers equations form an integro-differential equation for the evolution of the size distribution function $c(a, x, t)$

$$\frac{\partial c(a, x, t)}{\partial t} = V_0(a) \frac{\partial}{\partial x} \left[ c(a, x, t) - \int_0^\infty da' k(a, a') c(a, x, t) c(a', x, t) \right] +$$

$$\frac{\partial}{\partial x} \int_0^\infty da' D(a, a') \frac{\partial c(a', x, t)}{\partial x},$$

where kernel $k(a, a')$ is the continuous generalization of matrix $k_{ij}$ and diffusivity is a functional of $c$ with diagonal terms of zero and first order in $c$ and off-diagonal terms of first order in $c$. The solution of Eq(4.5) is well-defined due to the presence of $D$. It is noteworthy that for the kernels $k(a, a'), D(a, a')$ which dependence on $a$ and $a'$ consists of factorizable terms it may be more convenient to work with the Mellin transform of Eq(4.5) performed in variable $a$.

Generalization of Eqs(4.2), (4.3) to the continuous case is straightforward. We consider the simplest case of no ordering inversion. Then at any point $x$, on the interface (at late times) there exists maximal particle size $b(x)$ which is “eliminated” at this point. It is convenient to parametrize the spatial dependence by $b(x)$. Then concentration becomes a function of $a$ and $b$, i.e. $c(a, b)$ (and
certainly an implicit function of $x$ through $b(x))$. Particle hindered velocities are given by

$$v(a, b) = V_0(a)\gamma(a, b), \quad \gamma(a, b) = 1 - \int_0^b da' k(a, a')c(a'),$$

(4.6)

and the continuity equation reads

$$c(a, b)[v(a, b) - v(b, b)] = c(a, b - db)[v(a, b - db) - v(b, b)],$$

(4.7)

where

$$v(a, b - db) = V_0(a)\left\{1 - \int_0^{b-db} da' k(a, a')c(a, b - db)\right\},$$

(4.8)

Expansion of (4.6) - (4.8) to the first order to get an equation for $\partial c(a, b)/\partial b$ should be done carefully since $c(a, b)$ contains a singularity at $a = b$

$$c(a, b) = \frac{C(b)}{(b - a)^\mu(b)} + c^R(a, b),$$

(4.9)

where $\mu$ is positive, such that integral (4.6) converges, and $c^R(a, b)$ stands for the regular part at $a = b$. The exponent $\mu(b)$ can be related to other functions by using Eq(4.7), (4.8). A study not presented here shows that

$$\mu(b) = \frac{\partial \ln \gamma(a, b)}{\partial b} \Big|_{a=b},$$

(4.10)

i.e. concentration $c(a, b)$ becomes infinite at the point $a = b$. Given that the singularity is identified, an explicit differential equations of renormalization can be written for $dC/db$ and $\partial c^R(a, b)/\partial b$. 

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Numerical integration of Eq(4.5) enables to study time-dependent evolution of polydisperse suspensions. For this purpose we performed simulations on a Sun Sparc 2 workstation to fit the experimental data by Davis and Hassen [12], and Lee et al [14]. The simplest possible explicit difference scheme (Euler scheme) already works nicely for Eq(4.5), provided that spatial and temporal steps obey the conditions $\Delta x \ll D/V_0$, $\Delta t \ll \min(\Delta x/V_0, \Delta x^2/D)$. The results cease to depend on numerical resolution for $N > 30$ particle species (to model the size distribution function) and more than 1000 spatial points for each of the species given that the characteristic value of the hydrodynamic diffusivity exceeds $10^{-4} \text{cm}^2/\text{sec}$. Diffusivity of particles of size $a$ was selected to be

$$D_h = k'a_0V_0(a_0), \quad (4.11)$$

where $a_0$ is the average size. The results are presented in Figs. 4, 5 and 6. Fig.4 shows evolution of the size distribution function $c$ and resembles closely the numerical results by Davis and Hassen (see their Fig.2) if the latter are resolved in space. One can clearly see the appearance of singularities of the size distribution function. Their amplitude is restricted by diffusive resolution as we discussed above. Fig.5 compares simulation results with the experimental data for transmitted light intensity, which is given by [12]

$$\log[I(x,t)] \propto -\int \frac{da}{a}c(a,x,t), \quad (4.12)$$

scaled to the experimental amplitude range $I(x,0)/I(x,\infty) = 0.12$. The factor $k'$ was the adjustable parameter. It is interesting to note that we didn’t find any significant dependence upon the constant $k'$ for the values $k'$ up to about 10. Also
important is the observation that the interface width obtained at smaller $k'$ (we tried $1 \leq k' \leq 10$) is nicely comparable with the experimentally measured one. Smaller values of $k'$ require higher resolution or more advanced numerical schemes and were not attempted.

Our conclusion is different from the conclusion made by Davis and Hassen who used Eqs(4.2), (4.3) and discovered that in addition to the polydisperse width there is a diffusive-like contribution which they identified with the effect of gradient hydrodynamic diffusivity. We agree with this statement but point out that “bare” hydrodynamic diffusivity that enters Eqs(2.4), (4.5) doest not contribute to the interface width directly - it is achieved through the evolution of the equation. For instance, the distribution of the phase shifts influences the width. Thus, we think that the simulation of the coupled Burgers equation (4.5) is useful to decribe the results of the polydisperse sedimentation experiment. It should also be noted that the tails of the interface profile is sensitive to the real particle size distribution (which is not necessarily gaussian as assumed in Fig.5 and in Ref.12).

An analogous fit was performed for the data in Fig.2 of Ref.14. The fit is shown in Fig.6. Here the experimental standard deviation of particle size was only $2/3$ of that of Ref.[12], and polydispersity alone can not account for the observed width. With the help of hydrodynamic diffusivity (using $k' = 10$) we can get curves which are rather similar to experimental ones. The exception is the last curve, which position is not described by the motion with a constant velocity. We are thankful to Soonchil Lee who explained that the shape and position of this curve is affected by the presence of the lower interface between the suspension and sediment, and must be ignored in our analysis [31].
5. Thermal fluctuations

Now we discuss briefly the influence of intrinsic thermal noise on sedimentation which corresponds to Brownian motion of sedimenting particles and is only relevant at small Péclet numbers. One can further discriminate between two situations. In macroscopic fluid mechanics equilibrium thermal fluctuations have zero correlation length. Correspondingly, different particles experience different Brownian drag with amplitude reflecting their sizes. One may consider adding a delta-correlated (in size, space and time) gaussian noise $\xi(a, x, t)$ to the 3D generalization of Eq(4.5)(c.f. [7]). On the other side sufficiently small colloidal particles may be within the correlation of some thermal (especially Van-der Waals) fluctuations, then there is a correlated Brownian drag on particles of different sizes, and additive noise is no longer delta-correlated in particle size. This leads to complex behavior: for each noise-induced inhomogeneity a segregation of particles starts within initially uniform system. In the same manner as usual Burgers turbulence is an interplay between non-linearity and diffusion, we have here in addition the effect of size-coupling.

Even for the monodispersive system in thermal equilibrium with average concentration $c_0$ at temperature $T$ there are fluctuations, $c_1 = c - c_0$, which are described by Burgers equation,

$$\partial_t c_1 = V_0 \partial_x [(1 - 2kc_0 - kc_1)c_1] + D\Delta c_1 + \xi. \quad (5.1)$$

Here we again neglected the $c_1$-dependence of diffusivity, $\xi$ is the thermal noise with the correlator proportional to temperature. If one averages Eq(5.1) over the lateral cross-section of the sedimentation column, $\langle \ldots \rangle_\perp$, the resulting equation is no longer
a closed Burgers equation but their mean-field approximations coincide. The value of $h(x) = \int_{x}^{x} \left\langle c_1(x, t) \right\rangle_{\perp}$ is then analogous to the so-called interface height which was extensively studied in the framework of Burgers turbulence [32] and represents an example of dynamical critical phenomena. Nonequilibrium dynamics can be induced by changing the ambient temperature (liquid helium?) or switching off the interparticle interaction.

Another question is the influence of external thermal fluctuations when experiment is performed under incomplete thermostatic conditions. This issue was addressed experimentally [33] and discussed theoretically [7]. The detailed study of this effect which results are different from Ref.7 will be published elsewhere [11].

6. Conclusion

Application of (coupled) Burgers equation to describe sedimentation of (poly-dispersive) suspension and colloids enables one to reproduce experimental data and provides a basis for analyzing time-dependent sedimentation. We hope that this procedure will become a routine lab software and will help developing an industrial analysis for sedimentation processes.

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8. Figure Captions

*Fig.1* Fit of light intensity data versus tube height for a monodispersive suspension [13]. Curve 1 was taken after 195.2 hours after the beginning of experiment, curve 2 - after 374.7 hours. Experimental parameters used for the fit: \( a = 65\, \text{nm} \), \( c_0 = 0.009 \), \( \Delta \rho = 1.005 \, \text{g/cm}^3 \), \( V_0 = 0.12 \times 10^{-5} \, \text{cm/sec} \), \( T = 25^\circ C \), so that \( \eta = 0.0077 \, \text{g/cmsec} \), \( D_0 = 4.4 \times 10^{-8} \, \text{cm}^2/\text{sec} \). Both curves are scaled in vertical direction to meet experimental range of Fig.5 in Ref.13. This is the experiment at small Péclet numbers, so that hydrodynamic diffusivity is negligible with respect to the Brownian part.

*Fig.2* Numerical integration of two coupled Burgers equations (3.2). Parameters: \( V_1 = 0.1 \), \( V_2 = 0.05 \), \( k_{11} = k_{12} = k_{22} = 0.4 \), \( k_{21} = 0.35 \), \( D_1 = D_2 = 0.01 \). Initial conditions for \( c_1(x) \) and \( c_2(x) \) were selected to be their individual Burgers shocks in the absence of coupling with small shift with respect to each other, \( c_{-\infty} = 1 \), \( c_{+\infty} = 0 \) for both substances. Integration time \( t = 2000 \), \( \Delta t = 40 \) between profiles. \( c_1 \) is shown with lines, \( c_2 \) with broken lines. System size \( x = 100 \). The sign of velocities was selected such that the shock motion occurs from right to left. Initial and final profiles are shown with thick lines.
Eqs(3.2) do not impose any contraints on amplitudes of $c_{1,2}$. We can formally use the non-physical range $c_{1,2} > 1$.

**Fig.3** An example of complex temporal behavior. Parameters are the same as in Fig.2 except for $k_{21} = 0.4$.

**Fig.4** Snapshot of the solution $c(a,x,t)$ at time $t = 36 \text{ min}$ for the experiment [12]. Parameters: $a = 61(\pm 6) \text{ } \mu m$, $\Delta \rho = 1.384 \text{ } g/cm^3$, $c_0 = 0.05$, $\eta = 0.0085 g/cm sec$. With this parameters one obtains: $V_0(a) = 0.0141 \text{ } cm/sec$, $D_h(a) = k' \times 8.5 \times 10^{-5} cm^2/sec$. The value $k' = 1$ is used (see Fig.5). System length is 40 cm, initially gaussian size distribution with deviation $6 \mu m$ is used. Number of species $N = 26$, 1600 spatial points for each particle species.

**Fig.5** Fit of raw transmitted light intensity versus time for a polydisperse suspension (Fig.11 of Ref.12). The intensity is measured at heights $x = 1.5, 6.0, 24, 32 \text{ cm}$. Note that we use $k = 2.5$; this is the experimental value (!) which can be easily extracted from Fig.11 in contradiction with Table 1 (both from Ref.12). The other possible explanation is that Fig.11 was, in fact, taken for $c_0 = 0.02$, then $k \sim 5$ in agreement with Fig.12 of Ref.12 and Table 1. (In both cases the conclusion made in the text remains valid). The Richardson-Zaki dependence $(1-c)^k$ is employed instead of $1-kc$, here $c$ is the total volume fraction. The value $k' = 1$ is used to show that the hydrodynamic diffusivity is not required to account for the width of curves. At the top of theoretical curves one can notice small oscillations caused by insufficiently large number of particle species $N = 26$ when they get separated.

**Fig.6** Fit of volume fraction $\int da \ c(a,x,t)$ versus height $x$ for four different times: 332, 443, 567, and 1288 sec from left to right. Last experimental curve
at 2611 sec is not fitted since its average displacement is not described by a motion with constant velocity (see text). Experimental curves are from Fig.2 of Ref.3. Parameters: $a = 67.9(\pm4.0)\,\mu m$, $\Delta \rho = 1.57\,g/cm^3$, $\eta = 0.0164\,g/cm\,sec$, $c_0 = 0.1$. With these parameters: $V_0 = 0.0096\,cm/sec$, $D_h = k' \times 6.510^{-5}\,cm^2/sec$. The value $k' = 10$ is used. The Richardson-Zaki dependence $(1 - c)^k$ is employed with $c$ being the total volume fraction here.