# Effect of size polydispersity on granular materials

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We present particle dynamics simulations on fluidization of vertically vibrated granular beds where solidlike and fluidlike domains appear spontaneously. The existence of polydispersity in granular size significantly alters where the boundary between solidlike and fluidlike regions appears. A threshold value of polydispersity $\sigma_{th}$ exists above which the system becomes insensitive to the change in the degree of polydispersity. The value of $\sigma_{th}$ shows good agreement with the critical polydispersity reported on the solid-fluid phase transition in thermodynamic hard-core systems. The present result implies that the existence of polydispersity is crucial in some granular systems. [S1063-651X(96)06608-1]

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## I. INTRODUCTION

Granular materials often comprise a state with characteristics of both solids and fluids [1]. Especially, when granules are subjected to vibration many interesting phenomena appear, such as heap formation [2], convection [3], and size segregation [4]. Except for the studies on size segregation, theoretical investigations to date mainly treat monodisperse systems despite the fact that materials in experimental studies are inevitably polydispersed in granular size. To investigate the effect of polydispersity on fluidization of granules also have a strong implication in industry since vibrated beds are widely used in powder processing as a means of fluidizing and mixing granular materials.

Granules are generally treated theoretically as particles with contacting interaction and energy dissipation through friction. When the energy dissipation is negligible, theoretical treatment in statistical mechanics and thermodynamics should, in principle, give a good description of their motion and behavior. However, to apply methods of statistical physics to granular materials have been a challenging subject because of their shared properties of solids and fluids:regions where local density is low behave like fluids while high density domains behave like solids.

On the other hand, it is well known that systems of monodisperse hard-core particles exhibit thermodynamic fluid-solid transition with a coexistence region of two states in between [5]. In these hard-core systems, the phase diagram is dependent solely on density because hard-core potential only has two states with energy of zero or infinity. The temperature dependence only enters the partition function of these systems through the kinetic energy distribution.

Observation and comparison of the two systems mentioned above lead us to the hypothesis that some behaviors of granular systems consisting of particles sufficiently hard should be understood in an analogical context of thermodynamic hard-core systems [6].

Simulation studies of monodispersed granules subjected to vertical vibrations have shown that pressure and density waves propagate through the bed towards the bed surface [7]. In addition, it has been revealed that the profile of the kinetic energy per particle $E_k$ peaks at an intermediate height $h_{\mathrm{peak}}$. At bed heights $h<h_{\mathrm{peak}}$, granules show solidlike behavior since they are highly packed. In this region, $E_k$ increases along with the bed height $h$:as $h$ increases the density becomes lower and the particles become easier to move. At $h>h_{\mathrm{peak}}$, the behavior of the bed shows different properties in wave propagation. This region can be interpreted as a fluid phase of the granules. The value of $E_k$ goes through a minimum at the bed surface whereupon the value of $E_k$ increases drastically since the granules are bouncing freely on the top of the bed. The spontaneous appearance of the solidlike and fluidlike regions in these vibrated granules can be interpreted as a solid-fluid phase transition controlled by the local density of the granules.

Since it was found that the size polydispersity has a notable effect on thermal hard-core systems [8], it is speculated that the polydispersity also has an effect on the dynamic characteristics of granular systems. However, most of the theoretical studies on vibrated beds to date treat monodisperse systems which might not give a good description of polydisperse systems. Here, we investigate the effect of size polydispersity in vertically vibrated granular beds consisting of large and small particles of the same fraction. We show that the existence of polydispersity in granular size significantly alters the height profile of $E_k$ and the position where the boundary between the solid and fluidlike regions spontaneously appear. Concerning the effect of polydispersity, a clear analogy exists between this boundary in vibrated granular beds and the characteristics of solid-fluid phase transition in hard-core systems under thermal equilibrium.

## II. MODEL

Two dimensional particle dynamics simulations were performed for a model where the particles interact via a pairwise short-range repulsive force $\mathbf{f}_{ij}=-\partial\phi_{ij}/\partial\mathbf{r}_{ij}$ and a normal dissipation force $\mathbf{f}_i=-\gamma m(\mathbf{v}_{ij}\cdot\mathbf{r}_{ij})\mathbf{r}_{ij}/|\mathbf{r}_{ij}|^2$, where $\gamma$ is the dissipation coefficient [7]. The function $\phi_{ij}$ takes a form of

 

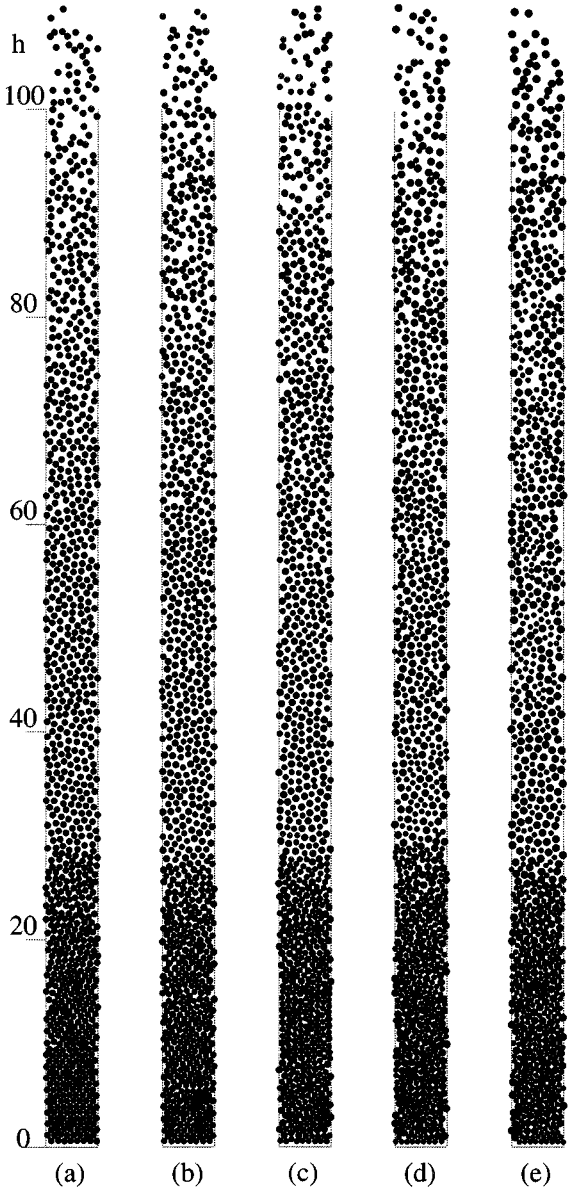


FIG. 1. Snapshots of the bed at $T=200$ for systems of $N=1000$, respectively, for $\sigma_0^2=$ (a) 0.0, (b) 0.0025, (c) 0.01, (d) 0.0225, and (e) 0.04.

$$\phi_{ij}=\begin{cases} \varepsilon\left[\left(\dfrac{d_{ij}}{\mathbf{r}_{ij}}\right)^{12}-\left(\dfrac{d_{ij}}{\mathbf{r}_{ij}}\right)^{6}+\dfrac{1}{4}\right] & \text{if } |\mathbf{r}_{ij}|<r_0, \\ 0 & \text{otherwise} \end{cases}$$

where $d_{ij}=(d_i+d_j)/2$ and $r_0=2^{1/6}d_{ij}$. The degree of polydispersity is expressed by $\sigma^2$, the variance of the distribution of particle diameters. Two kinds of particles with diameter $(1+\sigma_0)$ and $(1-\sigma_0)$ were mixed randomly at the same fraction, initially. Simulation studies starting from different distribution of particles were conducted. The mass of the particles is fixed to unity irrespective of the diameter. Energy is measured in units of $\varepsilon$. A periodic boundary condition was imposed in the horizontal direction. Sinusoidal vibrations of amplitude $A=3$ with vibrational acceleration $\Gamma=3g$, where $g$ is the gravitational acceleration, were applied to the system. Simulation studies of systems consisting of particle number $N=500$, 700, 1000, and 1400 in a simulation box with a width of 5 have been conducted. We set $\gamma=0.01g$. The system was vibrated for 100 to 800 cycles. The time $T$ is expressed in a unit necessary for one vibration cycle. Snapshots of system size $N=1000$ at $T=200$ for different values of $\sigma_0^2$ are shown in Fig. 1.

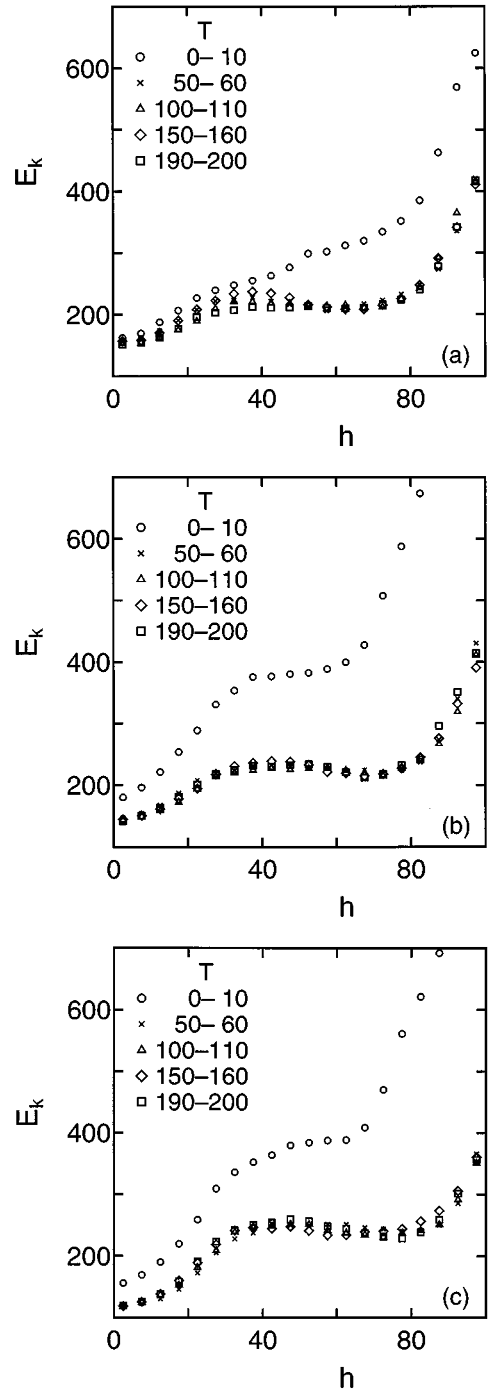


FIG. 2. Height profile of kinetic energy per particle $E_k$ of $N=1000$ at different times for $\sigma_0^2=$(a) 0.0, (b) 0.0025, and (c) 0.0225. Data averaged over ten vibrational cycles are used: $T=0$–10(○), 50–60(×), 100–110(△), 150–160(◇), and 190–200(□).

We observe the profile of the kinetic energy per particle $E_k(h;\sigma)$, which is a function also dependent on the degree of polydispersity $\sigma^2$, and analyze how the boundary between

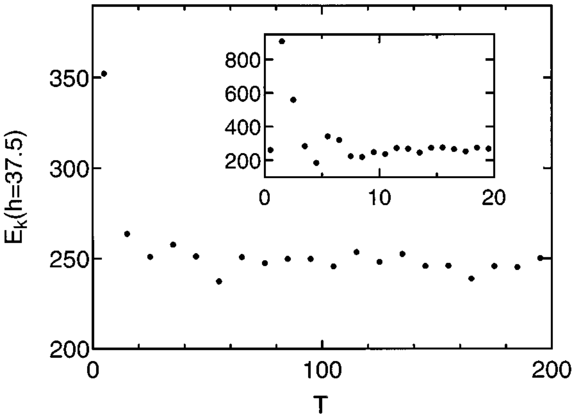


FIG. 3. Time evolution of kinetic energy per particle $E_k$ at $h=37.5$ for $\sigma_0^2=0.0225$ and $N=1000$. Data averaged over one (inset) and ten vibrational cycle(s) are used.

the solidlike and fluidlike regions is affected.

## III. RESULT

### A. Dynamics

First of all, we investigated the relaxation of the bed through the time evolution of the height profiles of $E_k$. Figure 2 shows the time evolution of $E_k(h)$ of $N=1000$ for $\sigma_0^2=$ (a) 0.0, (b) 0.0025, and (c) 0.0225. Data averaged over ten vibrational cycles are used. The value of each height is a mean value in the area of $5\times5$ around a given height. The height profiles peaks at an intermediate height ($h=40$), while the the bed surface lies at about $h=70$. Any systematic change in the fluctuation or relaxation of $E_k$ has not been noticed for changing $\sigma_0$.

As an example, we show the time evolution of $E_k$ for $\sigma_0^2=0.0225$ and $N=1000$ at $h=37.5$ where the fluctuation is large compared to $E_k$ at other heights, presumably because it is near the boundary between solid and fluidlike regions (Fig. 3). The values are an average of ten cycles for the main graph and one cycle for the inset. Figure 3 shows that $E_k$ is fluctuating around a certain value at $T>20$ and it can be said that $E_k$ is stable at $50\leqslant T\leqslant200$. [At the free surface region, it takes more time ($T\simeq50$) for $E_k$ to reach a stable state.] The time scale where $E_k$ is stable is well defined for $\sigma_0^2\leqslant0.0225$ at $T\leqslant200$.

### B. Kinetic energy profile

In this subsection we are concerned with the time scale where $E_k$ is stable as discussed in the preceding subsection. Figure 4 shows the value of $E_k$ against the bed height $h$ for various polydispersities for two system sizes (a) $N=1000$ and (b) $N=1400$. The height dependence of $E_k(h;\sigma)$ is qualitatively the same for all $\sigma_0$'s. From the bottom, $E_k$ grows until the peak around $h=40$ ($h=50$ for $N=1400$). At $h$ above the peak, $E_k$ decays for a while, and beyond the minimum around $h=70$ ($h=95$ for $N=1400$), it increases again. These three regions are interpreted as the packed-(solid), fluid, and free surface regions, respectively, as are studied in Ref. [7]. These maxima and minima of $E_k$ will become sharp when the height of the granular bed is increased [7]. When the system size is small ($N=500$ and 700), the boundary between the solidlike and fluidlike region become vague and the characteristic behaviors discussed below do not appear since the density near the boundary is below the solid-fluid transition density.

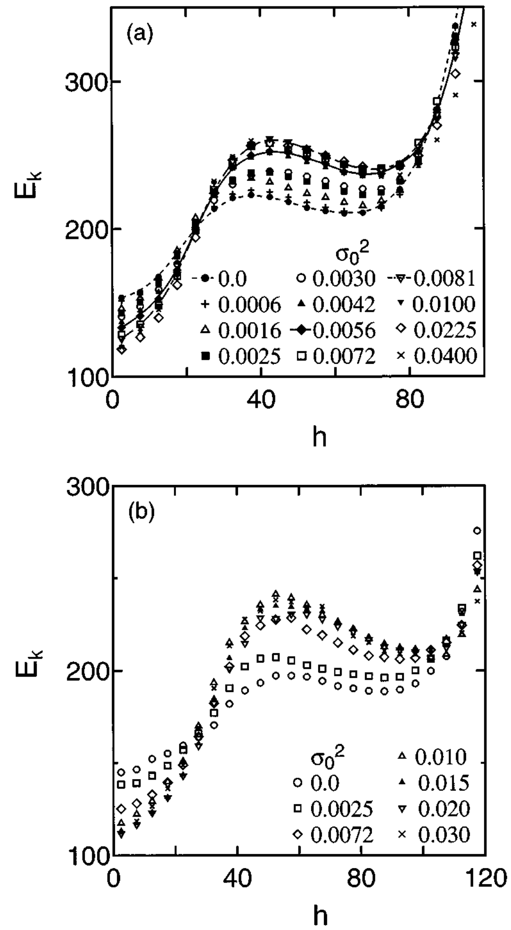


FIG. 4. Height profile of kinetic energy per particle $E_k$ for (a) $N=1000$ averaged over $T=50$–200 for $\sigma_0^2=$ 0.0(●), 0.0006(+), 0.0016(△), 0.0025(closed □), 0.0030(○), 0.0042(closed △), 0.0056(closed ◇), 0.0072(□), 0.0081(▽), 0.01(closed ▽), 0.0225 (◇), and 0.04(×), where best fit curves are drawn for $\sigma_0^2=$ 0.0(dashed line), 0.0056(solid line), and 0.0081(dot-dash line). An average value of two independent simulations starting from different initial configurations are used for all $\sigma_0$'s except for $\sigma_0^2=0.0042$, 0.0225, and 0.04 (average of three independent simulations) and for $\sigma_0^2=0.0016$ and 0.0072(single run). (b) $N=1400$ averaged over $T=50$–100 for $\sigma_0^2=$ 0.0(○), 0.0025(□), 0.0072(◇), 0.01(△), 0.015(closed △), 0.02(▽), and 0.03(×).

It can be seen from Fig. 4 that when $\sigma_0^2\leqslant0.0072$, the profile of $E_k$ is quantitatively sensitive to $\sigma_0$ and the maximum value of $E_k$ shifts toward higher values along with the increase of $\sigma_0$. The value of $h$ where $E_k$ reaches it's maxima also increases as the value of $\sigma_0$ increases. Note the drastic difference in $E_k$ for $\sigma_0^2=$ 0.0025 and 0.0072.

Figure 5 shows $E_k$ versus $\sigma_0^2$ at various bed heights for (a) $N=1000$ and (b) $N=1400$. Each data point for the same symbol corresponds to a independent simulation run. Focusing our attention to the fluidlike region (symbol ●), it can be clearly seen that $E_k$ is dependent systematically on the poly-

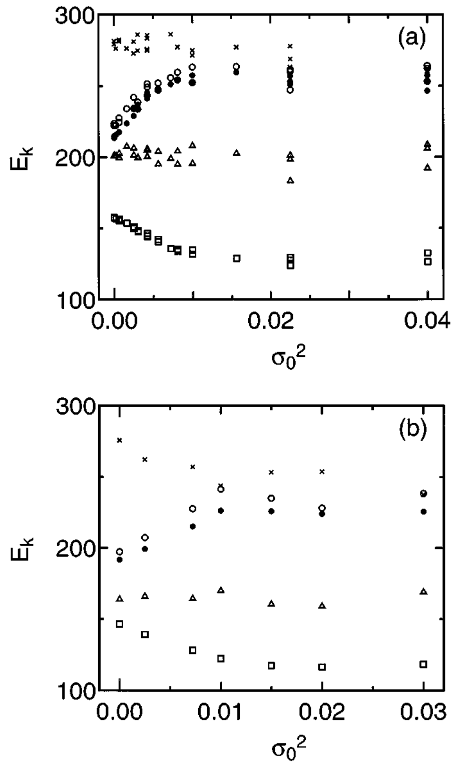


FIG. 5. The polydispersity dependency of $E_k$ for (a) $N=1000$, averaged over $T=50$–200 at bed heights $h=7.5$(□), 22.5(△), 37.5(○), 52.5(●), and 87.5(×), and for (b) $N=1400$, averaged over $T=50$–100 at $h=7.5$(□), 27.5(△), 52.5(○), 72.5(●), and 117.5(×).

dispersity when $\sigma_0^2<0.01$. For systems of $\sigma_0^2>0.01$, the values of $\sigma_0^2$ do not have any significant effect on $E_k$. Depending on whether $E_k$ is drastically affected by a slight change in polydispersity, $\sigma_0^2$ is divided into two behaviors, which will be referred to as the *insensitive* and *sensitive* behaviors to polydispersity. The threshold polydispersity is estimated to be $\sigma_{th}^2\sim 0.01$ for the present particle model. For systems of $\sigma_0<\sigma_{th}$, the energy profile $E_k(h;\sigma)$ depends on the polydispersity, while for systems of $\sigma_0>\sigma_{th}$, $E_k(h;\sigma)$ does not depend on the polydispersity. Similar polydispersity dependence of $E_k$ is recognized near the boundary between the solidlike and fluidlike regions (symbol ○). When $h$ is small (symbol □) there is also a sharp change in the behavior of $E_k$ at $\sigma_0<\sigma_{th}$, although the values of $E_k$ show the opposite dependence on $\sigma_0$: $E_k$ decreases as $\sigma_0$ increases until it reaches the value of $\sigma_{th}$. In the free surface region (symbol ×), where $E_k$ expresses the kinetic energy per particle moving freely (jumping out or falling towards the bed), a threshold polydispersity does not exist.

### C. Size segregation

It has also been studied whether segregation due to the size difference occurs in these polydispersed systems. Tendency of particle size segregation is observed in some of the present simulations, however it occurs in a different time scale compared to the relaxation time of $E_k$ discussed in Sec. III A. As an example, the height profile of the average diameter $\bar{d}(h)$ and the polydispersity $\sigma^2$ for $\sigma_0=0.0156$ and

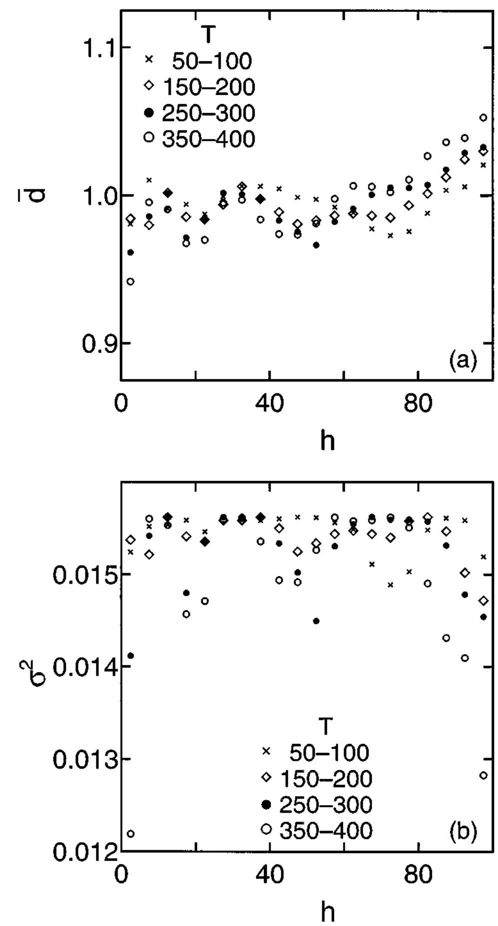


FIG. 6. Bed height dependence of (a) mean diameter $\bar{d}$ and (b) polydispersity $\sigma^2$ at $T=50$–100(×), 150–200(◇), 250–300(●), and 350–400(○) for $\sigma_0^2=0.0156$, and $N=1000$.

$N=1000$ are shown in Figs. 6(a) and 6(b), respectively. Indication of the beginning of the size segregation process, which might influence the polydispersity effect treated in the preceding subsection, is observed only at $T=350$–400. Thus the time scale $T\leqslant 200$ we use to claim the existence of $\sigma_{th}$ is free from the effect of size segregation.

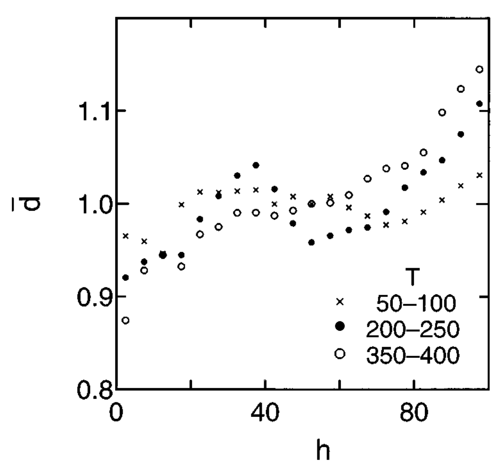


FIG. 7. Bed height dependence of mean diameter $\bar{d}$ at $T=50$–100(×), 200–250(●), and 350–400(○) for $\sigma_0^2=0.04$ and $N=1000$.

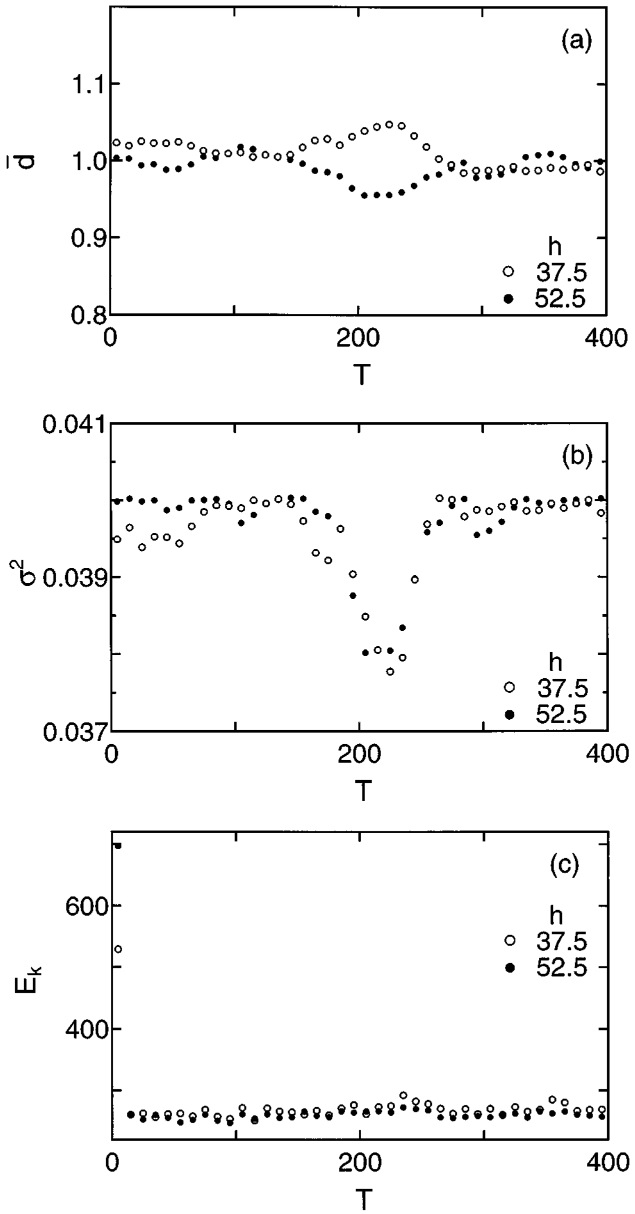


FIG. 8. Time evolution of (a) $\bar{d}$, (b) $\sigma^2$, and (c) $E_k$ at $h=37.5$(○) and 52.5(●) for $\sigma_0^2=0.04$ and $N=1000$.

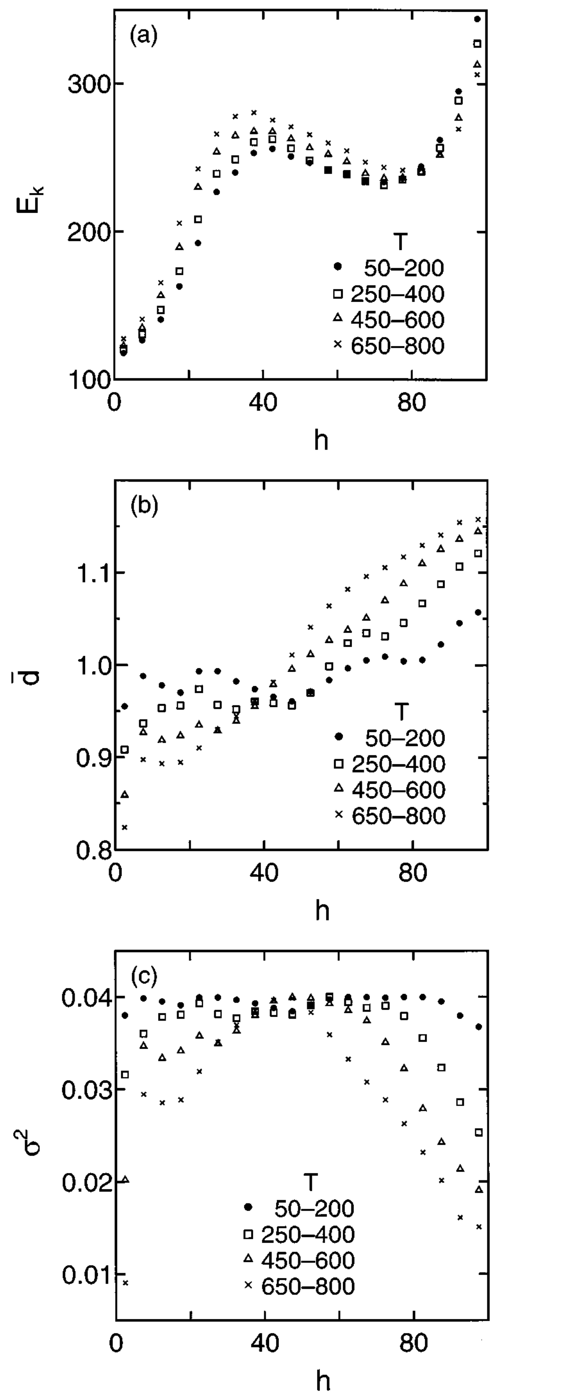


FIG. 9. Bed height dependence of (a) $E_k$, (b) $\bar{d}$, and (c) $\sigma^2$ for $\sigma_0^2=0.04$ and $N=1000$, at $T=$ 50–200(●), 250–400(□), 450–600(△), and 650–800(×).

Concerning the distribution of different sized particles, an interesting phenomenon has been observed which can be interpreted as local size segregation occurring at both the solidlike and fluidlike regions separately at an intermediate time scale. Figure 7 shows the height dependence of the averaged diameter $\bar{d}$ for $\sigma_0^2=0.04$ and $N=1000$ at $T=50$–100, 200–250, and 350–400. At $T=200$–250, there are two regions, around an intermediate height and the top of the bed, where larger particles dominate more than half of the granules in the area. The position of the maxima of $\bar{d}(h)$ at the intermediate bed height is slightly below that of $E_k$ in Fig. 4(a). This suggests that size segregation occurs at both solidlike and fluidlike regions of the bed and there exists a barrier at the boundary between two regions where larger particles are prevented to segregate to the top of the bed. This character of $\bar{d}(h)$ having a peak at an intermediate bed height is observed through $T=150$–300. Figures 8(a), 8(b), and 8(c), respectively, show the time evolution of $\bar{d}$, $\sigma^2$, and $E_k$ at $h=37.5$ and 52.5 for the same run. Since $h=37.5$ and 52.5 correspond to the maxima and minima of $E_k$ at $T=200$–250 (Fig. 7), the occurrence of local segregation in both solidlike and fluidlike regions at $150<T<280$ is clearly observed in Fig. 8(a) and 8(b). Such clear intercorrelation of the particle distribution above and below the boundary between the solidlike and fluidlike regions does not always appear; however the tendency that smaller particles accumulates above the boundary at intermediate time scales is generally observed. After 300 cycles of vibration, larger particles trapped in the middle of the bed start to penetrate the barrier and segregate towards the top of the bed. This process is recognized by the disappearance of the peak of $d(h)$ at the intermediate bed height at $T=350$–$400$ in Fig. 7.

When a system with high polydispersity is subjected to vibration for a long time, a clear tendency of size segregation appears. A drastic change in particle distribution will affect the profile of $E_k$: the values of $E_k$ drift slowly to higher values as the segregation process advance. Figure 9 shows (a) $E_k$, (b) $\bar{d}$, and (c) $\sigma^2$, versus bed height $h$ for system of $\sigma_0^2=0.04$ and $N=1000$ at different times. Each symbol refers to data averaged over 150 vibrational cycles. Figure 9 clearly shows that as the segregation process advances and large local differences in polydispersity appear the values of $E_k$ will shift towards higher values most drastically near the boundary of the solidlike and fluidlike regions.

## IV. DISCUSSION

The effect of polydispersity in granules under continuous vertical vibration has been investigated. The kinetic energy profile $E_k(h)$ relaxes quickly compared to the particle movement leading to size segregation which makes it possible to investigate the effect of polydispersity free from the effect of segregation. A boundary between high and low density domains, which was observed for monodisperse systems, appears as well for polydisperse systems. We have shown that there exists a threshold value $\sigma_{th}$ in polydispersity which divides the ''sensitive'' and ''insensitive'' behaviors of the bed: When the polydispersity is under $\sigma_{th}$ the value of $E_k$ is sensitive to a small change in $\sigma$, while $\sigma>\sigma_{th}$, the change in $\sigma$ does not have any significant effect on $E_k$.

The value of $\sigma_{th}$ is rather small compared with the experimentally and industrially relevant values of polydispersities. So the present result implies that the introduction of the polydispersity might be crucial in some theoretical and simulation studies of granular systems since polydispersity can modify the behavior of the system quantitatively.

As mentioned in the introduction, thermodynamic monodisperse hard-core particle systems show fluid-solid transition with a coexistence region when the density is controlled [5]. Furthermore, it has been shown that size polydispersity changes the feature of the fluid-solid transition under thermal equilibrium [8]. Introducing small polydispersities into these systems shifts the phase transition to higher densities and narrows the coexistence region. Above a critical polydispersity $\sigma_c$, the coexistence region vanishes. The critical polydispersity was estimated to be $\sigma_c^2\simeq 0.008$ of the average size, where $\sigma$ denotes the standard deviation of the size distribution function [8]. Above $\sigma_c$, the properties of the transition and of the phase at densities higher than the transition density are different from those below $\sigma_c$, and show characteristics of glass transition [9].

The existence of the sensitive and insensitive behaviors to polydispersity in vibrated granules can be understood in the analogy of the above mentioned behavior of thermodynamic hard-core systems. The value of polydispersity where the coexistence region in the solid-fluid transition disappears is approximately the same as the threshold value dividing the sensitive and insensitive behaviors in this study. This coincidence supports our hypothesis concerning the analogy of granular systems with thermodynamic systems. Local size segregation effectively decreases the polydispersity and this is presumably the reason why $\sigma_{th}$ concerning the sensitive and insensitive behaviors is slightly higher than $\sigma_c$ the thermodynamic prediction along with the effect of the particles being softer than the hard-core model [10].

We have succeeded to observe the effect of polydispersity on granular materials through the kinetic energy profile in vertically vibrated beds. There exists a time scale where the kinetic energy is stable which is effective in observing how the degree of polydispersity changes the total behavior of the granules free from the effect of size segregation. There is a threshold value in polydispersity $\sigma_{th}$ which divides the sensitivity of the granular bed against the change in $\sigma$. The analogy between the existence of critical polydispersity $\sigma_c$ in thermodynamically equilibrium systems around the solid-fluid transition and $\sigma_{th}$, studied in this paper, suggests that the polydispersity effect will appear not only in vibrated beds, but also in many other granular systems. It is expected that the existence of polydispersity gives an unnegligible effect to the behavior of granular systems.

## ACKNOWLEDGMENTS

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