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Influences of the interstitial liquid on segregation patterns of granular slurries in a rotating drum

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Granular mixtures immersed in a liquid (slurries) show segregation dynamics which are quantitatively and qualitatively different from those of dry systems. The principal mechanisms of the segregation dynamics in slurries, as well as the relevant material parameters that must be taken into account in a dynamic description are not sufficiently understood so far. We investigate experimentally the influence of the viscosity of the interstitial liquid on the coarsening of axial segregation patterns in a horizontally rotating mixer. It is found that not only the characteristic time scales but also fundamental structural features of these patterns are influenced by the viscous properties of the liquid component.

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I. INTRODUCTION

Segregation processes in mixtures of granular materials are encountered in everyday life as well as in various technological processes [1–3]. Even though engineers have developed efficient ways to handle such materials, many fundamental questions in these systems are still poorly understood. A classical example is the formation of segregation patterns of granular mixtures in a horizontally rotating drum. The geometry of this system is particularly simple. Its preparation is easy and observation techniques are straightforward.

Axial segregation has first been described by Oyama [4] already more than 60 years ago. Since then, the rotating drum experiment has kept attracting tremendous scientific interest, particularly during the last decade. Experiments have been performed in different geometries and with a variety of materials (e.g., Refs. [4–28]), and an abundance of experimental data has been provided. Theoretical studies include numerical simulations and the development of analytical models [28–41]. A more detailed introduction into earlier studies concerning the rotating drum experiment has been given elsewhere [25].

We consider here the classical case of a long horizontally rotating drum partially filled with a granular mixture. The granulate consists of a binary mixture of spherical beads of the same material (glass) with same densities but different sizes. The rotation is sufficiently slow so that the granulate is fluidized only in a shallow surface layer.

The segregation phenomena observed in such a horizontally rotating long mixer, initially filled with a well-mixed granulate, can be roughly classified into three stages: Already during the first few rotations of the mixer one observes a radial segregation [42] of the material. Small sized particles are enriched in an axial core of the granular bed, while the large size particles accumulate in the outer region. When the rotation speed of the drum is in the appropriate range, a second type of segregation pattern can form: The core structure becomes unstable and develops an axial modulation, its wavelength being roughly of the order of magnitude of the drum diameter. During a period of the order of 100 rotations, this process leads to the formation of an axial band pattern [14,15,27]. Along the drum axis, regions with more or less perfectly segregated smaller particles alternate with regions where an axial core of small particles is surrounded by segregated larger particles. These segregation bands are easily identified optically. The range of rotation frequencies where this axial segregation process is observed depends upon several details of the material composition and the experimental geometry. Usually, this band pattern is not stable, but a gradual coarsening of the structure sets in. This process occurs on a time scale of several 1000 rotations of the mixer [23]. The long-term evolution can finally lead to a complete demixing of the granulate into two axial compartments. One of them contains only small size particles, the second one is formed by the larger particles which surround a core structure of small size particles. We note, however, that a systematic investigation of the final state of segregation in the rotating drum has not been performed so far. The spontaneously formed axial band patterns can be very robust and persistent under certain experimental conditions. Experiments have shown that at appropriate rotation rates, an array of alternating segregation bands is practically stationary, stripes in the center of the pattern survive several 10,000 rotations of the mixer without noticeable changes. In a drum of finite length, such patterns dissolve stripe by stripe from one or both lateral ends due to boundary effects [25].

A basic distinction between two types of experiments must be made regarding the interstitial medium. Most experiments so far have been performed in dry systems where the granulate is immersed in air. Few experiments [22–25,43,44] deal with granulates immersed in a liquid of lower density (slurries). Dry granular mixtures and slurries are of equal importance in technological processes. However, they differ essentially in the mechanisms that are relevant for their microscopic descriptions. The features that distinguish slurries from dry granulates have not been studied systematically so far. A comparative investigation of dry and wet systems containing the same granulate has been presented by Fiedor and Ottino [23]. It has become evident from their measurements that there are many qualitatively similar features of both systems, like the very formation of the axial segregation structures at certain rotation velocities of the container, the general composition of these structures and the coarsening processes. A logarithmic decay law for the number of bands in the granulate has been described by these authors [23]. They have measured the time dependence of the number of stripes in square and circular drums containing dry and wet granulates. In these experiments, the evolution of the stripe pattern was observed for a few 1000 rotations, where coarsening leads to the dissolution of approximately one-half of the initial stripes. There was no clear difference in the decay constants for dry and wet systems. The parameters of the decay curve (versus number of rotations) were almost the same for both systems in the circular drum geometry, and they differed by almost a factor of 2 for the square tumbler. From these results one may conclude that the differences between dry and wet mixtures in one and the same geometry are no more important than the differences between the two geometries for one and the same mixture.

On the other hand, the parameter range for the rotation speed at which the axial segregation phenomena are observed is shifted, the wet systems are spun considerably slower [23]. It seems also that some detailed features like the formation of traveling waves are more pronounced in the dry systems. However, there is no clear concept about the role of the interstitial fluid so far. The bulk liquid is in motion relative to the granular bed and to the glass walls of the tube. This fluid flow may influence the motion of individual beads in the fluidized layer of the granulate. However, the redistribution processes during the coarsening of a segregation pattern are very slow on the time scale of the drum revolution, so that a direct connection between the fluid flow and the pattern dynamics is not evident. Another influence of the fluid, capillary forces between granular beads, would only be effective when the granulate were not completely immersed in the liquid. This is not the case in the systems studied here. One further effect of the interstitial liquid is that the collisions between the particles are damped stronger than in the dry granulate. Finally, the granular material experiences a buoyancy force in the liquid so that the particles appear specifically lighter in slurries. So far, it has not become clear how these effects must be incorporated into models for granular mixtures, for example, in molecular dynamics simulations. Summarizing this, one may conclude that the two essential physical parameters that determine the influences of the fluid environment in slurries are the viscosity of the liquid, controlling dissipation of the mechanical energy, and its density, controlling the effective weight of the granular components. One may expect, however, that the buoyancy effects are small as long as the variation of the density of the liquid is small compared to the density difference between fluid and granulate. In this study, we modify the viscosity of the interstitial liquid in segregation experiments with slurries. We replace water by different water-glycerol mixtures. The influences of the increased viscosity are analyzed by comparing the experimental observations with a system where pure water has been the interstitial liquid [25]. In order to exclude that density differences between the pure water and the water-glycerol systems are relevant, we test the influence of density changes in experiments with a salt solution.

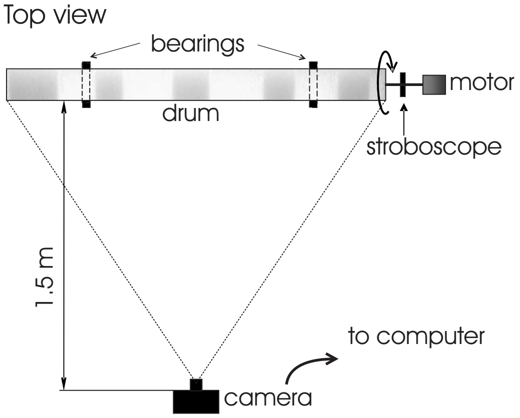

FIG. 1. Sketch of the experimental setup (not to scale). The drum rests on two ball bearings on each side, it is driven by a dc motor. The rotation speed is monitored by a stroboscopic counter. The drum is illuminated from the top. A digital camera at the same height as the drum, positioned in a distance of 1.5 m, automatically takes photos in regular intervals. The width of the drum shown is 662 mm. Typically, observation times ranged from 1 day to 1 week.

II. EXPERIMENT

The experimental geometry is sketched in Fig. 1. We use a setup that has been described earlier [25]. It consists of a cylindrical mixer driven by a dc motor with a stroboscopic disk for rotation velocity measurements, an illumination source and a computer controlled camera that allows to take series of images in defined time intervals. The mixer consists of a Duran glass tube (*Schott*) with $d=36.8$ mm inner diameter and 662 mm length. The fill level of granulate was 1/2, i.e., the dry mixture occupies one-half of the drum volume. After loading the granulate, the drum is filled up with the liquid. The mixer is closed tightly at both ends, with an aluminum plug on the motor side (right-hand side in the images), and a teflon plug on the opposite side. Rotation speeds in our experiments are moderate, Froude numbers $R\omega^2/g$ (with the angular velocity ω of the drum, gravitational acceleration g, and inner tube radius R), giving the ratio of inertial forces and gravitation forces on the granulate, are much smaller than 1. The Froude number is independent of the properties of the embedding liquid.

In a previous experiment with this setup, we have studied granular mixtures that were completely submerged in plain water [25]. The same granulate is used in the present study. The mixtures consist of transparent spherical glass beads with bimodal radius distribution. The radii of large and small particles were approximately 3:1, with an average diameter $d_L=1.5$ mm of the large spheres, and $d_S=0.55$ mm of the small spheres. The large size beads are practically monodisperse, the radius distribution of the small size beads was between 0.50 mm and 0.63 mm. The initial mixtures were prepared with a 50%:50% ratio of both species (tapped volume). We use the data from the earlier study of the pure water system [25] as a reference and study the same mixtures in glycerol-water mixtures and in a solution of sodium iodide in water. The parameters for the different liquids are com-

TABLE I. Compositions, densities and kinematic viscosities of the interstitial liquids.

Liquid	Density (g cm^{-3})	Viscosity (23 °C) (mm^2/s)
H_2O (pure)	1.0	0.995
H_2O +23.1 wt. % NaI	1.17	0.913
H_2O +40 vol % glycerol	1.11	3.58
H_2O +80 vol % glycerol	1.19	19.6

piled in Table I. The densities and kinematic viscosities have been determined in independent experiments with commercial viscosimeters and density meters.

The motivation of replacing the water by glycerol-water mixtures of different composition was to study the role of the viscosity of the liquid on the pattern dynamics. With the two different concentrations employed in this study, the viscosity could be varied by a factor of about 20. Experiments with higher concentrations of glycerol ($>90\%$) turned out to be unsuccessful, since no pattern formation was observable in reasonable observation times, certainly as a consequence of the much higher viscosities of these mixtures (up to 1000 times that of pure water). Since the glycerol mixtures have a slightly higher density than water, one cannot exclude *a priori* that the different buoyancy forces respective to the pure water system play a role in the granulate dynamics as well. Therefore we have prepared a solution of sodium iodide which has a density comparable to the higher concentration glycerol mixture but has a viscosity rather close to that of pure water. This allows us to distinguish between density and viscosity effects, provided that our assumption is justified that no other physical properties of the liquid are relevant for the granulate dynamics.

Special care has been taken to avoid demixing of the granulate during filling of the tube. A good criterion for the quality of the initial mixture is the formation of a pattern of uniformly distributed bands along the axis of the container during the axial segregation.

The optical image of the granulate in the mixer is basically determined by scattering of light at the particle-liquid boundaries. Individual grains are not resolved in the images taken by the camera. Initially, the tube appears uniformly gray. After axial segregation, the regions consisting of smaller beads appear darker in the images. Regions consisting of larger beads appear brighter. Even with a core structure of small beads inside these bands, these regions are clearly distinguishable from the bands of small particles. The core is not directly visible in our optical geometry, its existence may influence the brightness of the bands of large particles to a small extent. Nonperfect segregation results in a decreasing contrast between the bands of both types. This is discussed in more detail below. A typical image is shown in Fig. 2. The drum rotates such that the upper part moves away from the camera, thus the slope of the granulate inside the mixer is on the back side. We evaluate the images in the following way: From each image in the sequence, an axial intensity profile is constructed. We choose a horizontal cross section approximately in the middle of the tube image, and a vertical range of a few millimeters is averaged for each datum point in that profile. The time intervals between the images were chosen in relation to the rotation rate of the mixer, typically pictures were taken every 20 rotations or faster. We choose a compromise between sufficient temporal resolution and manageable data volume. A much larger image frequency is in principle possible, but would not add essential further information. The profiles of successive images are stacked to space-time plots (see Figs. 3–9).

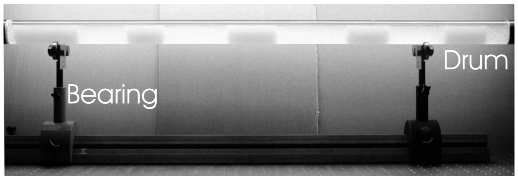

FIG. 2. View of a typical experiment, granulate of 1:3 (radius ratio) spherical beads in pure water. The dark bands are composed entirely of small sized particles, the bright bands are rings of large size particles around a narrow core of small sized beads.

III. RESULTS

Before the decay of the segregation band patterns is analyzed quantitatively, we compare the spatio-temporal plots in Figs. 3–9 on a qualitative level. The first image, Fig. 3, has been recorded with pure water as interstitial liquid. It has been recorded at a speed of 15 rpm and it represents the standard coarsening scenario which is observed in this system at rotation rates between approximately 10 and 20 rpm.

The glycerol systems (40% and 80%) develop patterns at considerably lower rotation speeds. Regular band patterns appear in the 80% glycerol mixture already at 2.5 rpm, see Fig. 4. Regular bands in the 23.1 wt. % NaI solution are found only at rotation speeds above 10 rpm, in the same range as in the pure water system. In the glycerol mix as well as in the NaI solution, some basic features of the pure water

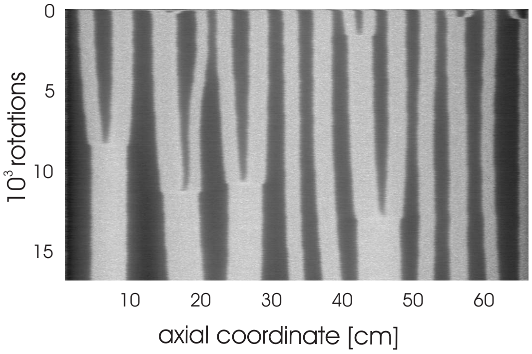

FIG. 3. Coarsening of the granular stripe pattern in a 662 mm long drum filled with pure water, rotation speed 15 rpm. The space-time plot is constructed from axial cross sections of the tube at the position of the tube center.

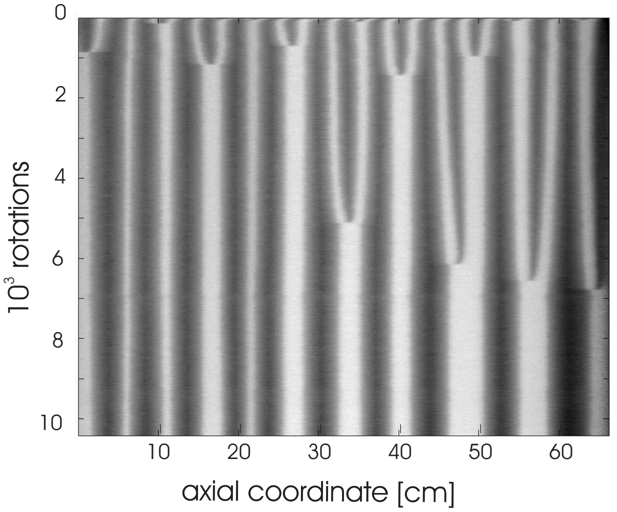

FIG. 4. Coarsening of the granular stripe pattern in a 662 mm long drum filled with 80% glycerol in water, rotation speed 2.5 rpm.

system are reproduced. Bands of large particles are completely separated from each other and the bands of small particles represent impermeable barriers for the beads of the larger species. Thus, all dynamics is restricted to the redistribution of the small beads [25]. However, the fast spun high viscosity mixtures form a certain exception to this. In these samples (see Fig. 7), the segregation is far from being complete and the large beads can penetrate the bands of small beads. Thus, the dynamics is qualitatively different, both species are involved in the pattern dynamics there. This feature is discussed in detail below.

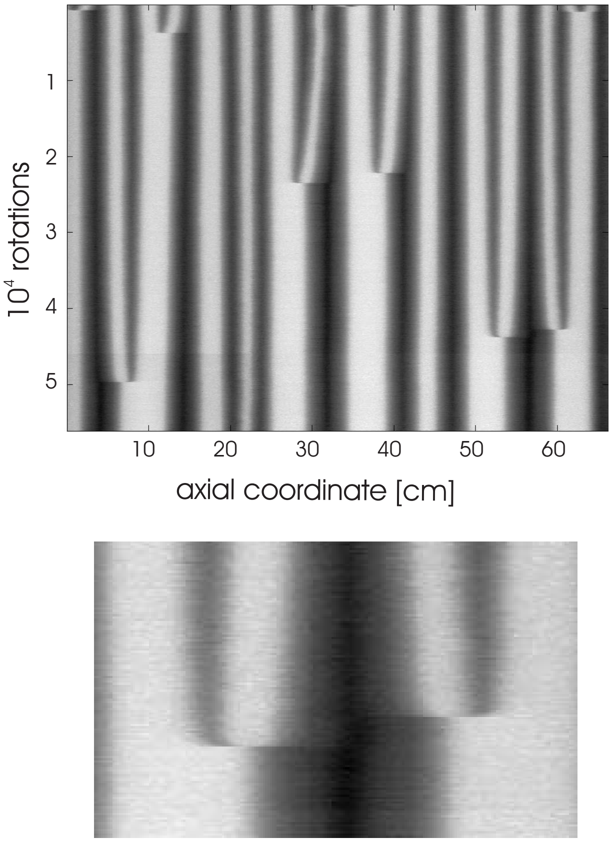

FIG. 5. Coarsening of the granular stripe pattern in a 662 mm long drum filled with 40% glycerol in water, rotation speed 10 rpm. The bottom image shows an enlarged detail of a 15 cm by 1000 rotations region in the bottom right-hand part of the top image, constructed from a sequence of 50 profiles.

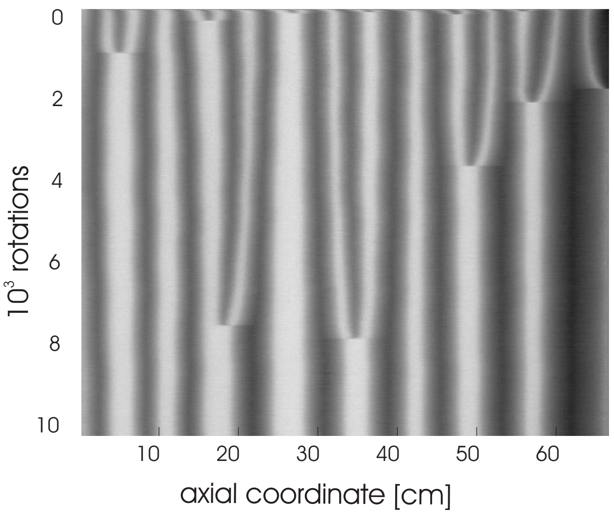

FIG. 6. Coarsening of the granular stripe pattern in a 662 mm long drum filled with 40% glycerol in water, rotation speed 20 rpm.

Experiments performed with the NaI solution (Figs. 8 and 9) produce results that are qualitatively and quantitatively comparable to the pure water experiments. They indicate that the features described above for the glycerol mixtures are not a primary consequence of the different fluid densities of the glycerol and pure water systems (the NaI solution has a density that is similar to the higher concentration glycerol mixture). The effects reported for the glycerol mixtures must be attributed exclusively to a higher fluid viscosity.

Another striking difference between the high and low viscosity samples is the internal structure of the segregation bands. In the images of the glycerol mixtures, the brightness of the darker bands (small beads) decreases gradually towards the centers of the bands. This is evident in particular in the profiles in Fig. 7. In contrast, bands of the small beads in the pure water system (Fig. 3) are optically rather homogeneous. This will become more evident in the quantitative comparison of intensity profiles [see Figs. 10(d, e)].

Although we have no direct quantitative relation between the local composition of the granulate and the optical intensity in the profile, the gradual transition seen, e.g., in Fig. 7 is strong evidence for a gradual change of the granulate composition in the axial segregation pattern over regions of several centimeters. The bottom graph of Fig. 7 shows two selected optical profiles from the top picture. It compares a profile at an early stage of the coarsening process with the profile at a later stage. The profile constructed after 80 000 rotations shows that the contrast between the two types of bands is much weaker than in the later profile. This indicates that the particles are poorly segregated, the small bead bands contain a considerable share of large beads and vice versa, the regions of large beads are mixed with a certain amount of

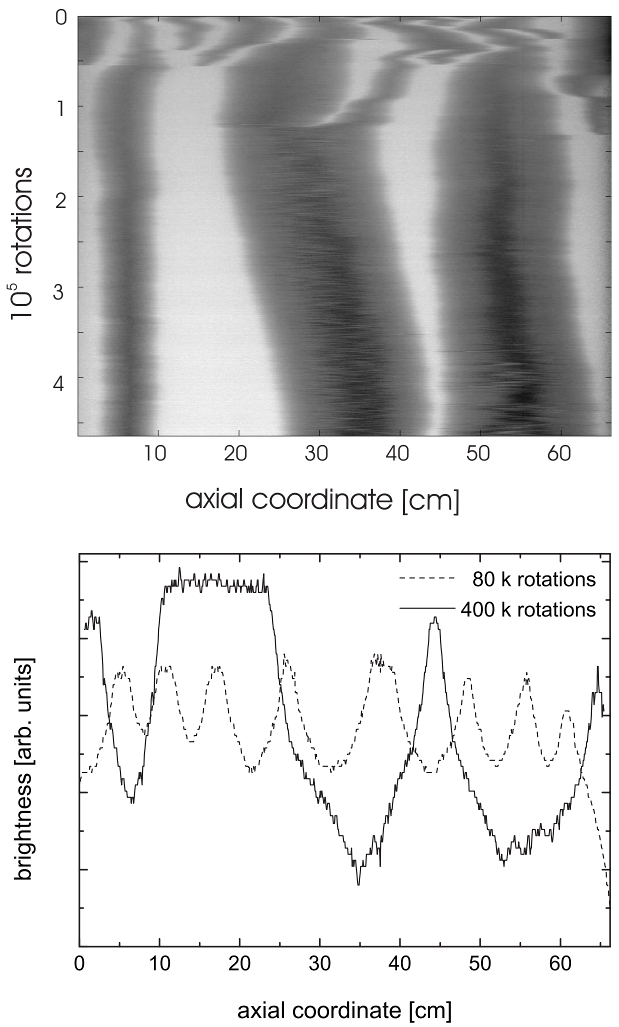

FIG. 7. Coarsening of the granular stripe pattern in a 662 mm long drum filled with 40% glycerol in water, rotation speed 30 rpm. The bottom image shows the optical intensity along the axial coordinate, after 80 000 rotations and after 400 000 rotations at 30 rpm. It is apparent that the segregation is not complete and that the large particles penetrate the zones formed by the small particles at this rotation speed.

small beads. The profile taken at the later stage (400 000 rotations) gives some indication that the saturated upper plateau corresponds to a band where large beads surround an axially uniform core structure of small beads. The lowest intensity regions correspond to segments with maximum concentration of small beads in the radial cross section. The transition ranges where large beads mix into the bands of small size beads (reflected in the slopes in the optical profile) extend over distances of the order of 100 mm. In this spatiotemporal plot recorded at 30 rpm (the fastest rotation experiment that we have performed with the 40% glycerol mixture) the segregation is so incomplete that the bands of small size beads are no longer impenetrable barriers for the large component. This refers in particular to the initial phase ($\approx 10^5$ rotations) where these bands are only a few centimeters broad. The large beads diffuse between adjacent bands, which is reflected in the continuous change of the width of large bead bands with time (Fig. 7). This feature distinguishes this particular experiment from the experiments performed with lower viscosity fluids and with lower rotation velocities. In the pure water system it has been established by MRI experiments [25] that even narrow bands of small beads are fully segregated, i.e., they do not contain any beads of the large species. Therefore, these bands inhibit the redistribution of the large species. As a consequence the bands of

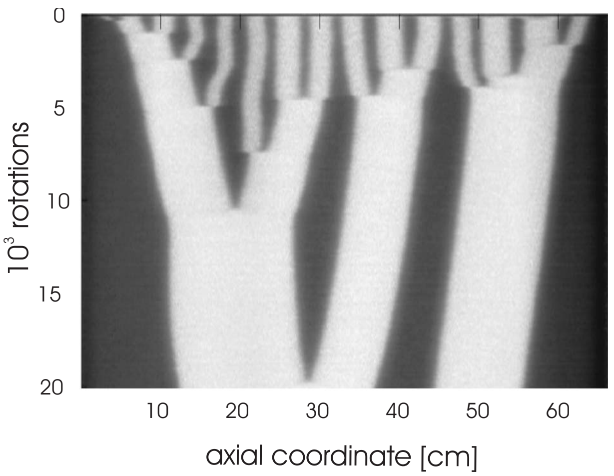

FIG. 8. Coarsening of the granular stripe pattern in a 662 mm long drum filled with a 23.1% NaI solution in water, the rotation speed is 20 rpm.

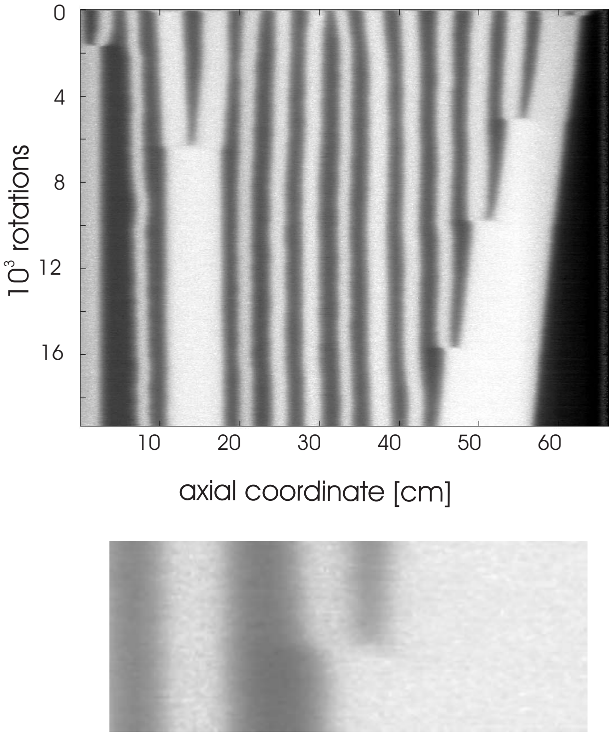

FIG. 9. Coarsening of the granular stripe pattern in a 662 mm long drum filled with a 23.1% NaI solution in water, rotation speed 30 rpm. The detail image at the bottom shows the evolution of a typical 15 cm section during 1000 rotations, constructed from a sequence of 50 profiles.

large particles cannot shrink or grow. They may shift axially but keep constant widths unless they merge completely with a neighbor band.

The band positions in Fig. 7 stabilize only after several 100 000 rotations, after sufficiently broad stripes have formed and the centers of the small particle bands in these patterns are nearly free of large beads. The bands of small size particles at the progressed state of coarsening finally seem to become large enough to block the passage of the large beads. Since these observations are typical for the glycerol mixtures but not for the NaI solution (see next paragraphs) nor for pure water, we conclude that the viscosity of the fluid plays the decisive role for this softening of the band structure.

In the glycerol mixtures, a continuous gradient in the optical profiles indicates a gradual transition from the well segregated small particle region to the band of large particles. This feature is more pronounced when the rotation speed is increased and when the liquid has a higher viscosity. The boundaries of the individual bands are much sharper in the experiments with low viscosity fluids (Figs. 3, 8, and 9).

The detailed optical appearances of selected single bands in low and high viscosity surroundings are compared in Fig. 10. In the less viscous NaI solution (a), the boundaries between the bands of small and large size particles are sharp. The segregation of the two types of particles is complete. The band of small beads has a rather uniform brightness in the axial direction. The same applies to a comparable structure in pure water, (c). In contrast, the segregation pattern in the high viscosity liquid (b) shows the gradual brightness change in axial direction that has already been described in the context of Fig. 7. Figure 10(d) compares the axial profiles of images (a) and (b) quantitatively. While the optical contrast in axial direction drops across roughly 10 mm in the NaI sample, the slope is practically continuous along the small bead stripe in the glycerol sample. One must conclude that there exists a continuous segregation gradient in the fast spun glycerol samples. Figure 10(e) shows two selected profiles of the water system for comparison. The bands consist of a central region where the optical intensity changes by only a few percent, and side regions with a steep drop in optical contrast. It is also evident that after comparably few rotations of the water system (as compared to the glycerol mix profiles in Fig. 7, bottom), the optical contrast of the bands has almost reached its final value. We note, however, that the evaluation of the optical profiles is but a qualitative indication for the segregation structures. Since light is multiply scattered, narrow stripes of small beads always appear slightly brighter than broad stripes, and in particular, the details of the optical profiles must not be overestimated. Nevertheless, the differences between the low and high viscosity samples are unambiguous.

The MRI image of a stripe in the pure water system, Fig. 11, demonstrates clearly the perfect segregation of the small beads (see also MRI data in Ref. [25]). The image shows a stripe that is approximately 28 mm wide, at the sides one can see the core of small beads extending into the bands of large beads. MRI experiments with the glycerol mixture have not been successful, therefore the only evidence for the uncomplete segregation is the comparison of the optical contrast data.

Another characteristic feature observed in the glycerol mixtures, Figs. 4–7, which at first glance seems to be at least quantitatively different from the pure water system, is the dissolution scenario of individual bands. As one can see in the detail image of Fig. 5, the stripes disappear abruptly. There seems to be some minimal stripe widths of roughly 10 mm at which individual bands of small beads are able to survive (at least temporarily). Thicker bands may gradually lose material to the neighboring stripes (over periods of several 100 or 1000 rotations), but as soon as a stripe has reached the critical width, it collapses: Within a few dozen rotations, the material of the vanishing stripe is completely absorbed by the neighboring stripe(s). Figure 12 shows a selection of optical profiles for the last 850 rotations before and about 1000 rotations after such a collapse. The profiles have been extracted from the pattern in Fig. 5, after about 42 000 rotations, when the stripe at position 52 cm collapses. There is little dynamics of the dark band at ≈52 cm within a period of several 1000 rotations before the collapse, except for a small decrease of the stripe width and a slight shift towards its right-hand neighbor. Then, during a period of less than 40 rotations, the band is extinguished completely and the material has completely been relocated to the right-hand side, over a distance of roughly 10 mm. It is added to the neighboring stripe (compare the two solid graphs in Fig. 12, immediately before and after the collapse). For the following (at least 1000) rotations, there is again only little dynamics of the coarsened pattern. This form of collective redistribution of material in the collapse of individual stripes seems to be singular for slurry systems, and it is particularly well developed in a high viscosity liquid environment. In pure water and in a sodium iodide solution, the scenario of fast collapses of dissolving bands is also indicated (see, for example, Fig. 8), but since the dynamics of the stripes is equally strong before the collapse, it is not as obvious as in the higher viscosity glycerol mixtures where the bands remain almost stable before their sudden collapse.

After these qualitative descriptions, we discuss the coarsening for both types of fluids on a quantitative level and compare them to the pure water system. Figures 13 and 14 show the evolution of the number of stripes in the glycerol and NaI systems, respectively. The thick solid line is a graph taken from the average of all data available for the pure water system with the same granular mixture at rotation speeds between 10 and 20 rpm [25]. This is the range where in the pure water system, the standard coarsening scenario is found, i.e., the band system is gradually destroyed by a random dissolution of individual stripes.

The initial number of stripes per tube length developed during the axial segregation is comparable in all systems, there is only a slight trend to somewhat smaller values for the low viscosity (NaI) system and somewhat higher values for the high viscosity (glycerol) system, but the differences are well within the statistical distribution width. Further we note that, apart from the fact that the stripes appear at much lower rotation speeds in the glycerol system (in particular in the 80% glycerol solution), the decay of the number of stripes in dependence upon the number of rotations is in the initial stage not very sensitive to the liquid surrounding. When one looks at the long term data, the behavior of the

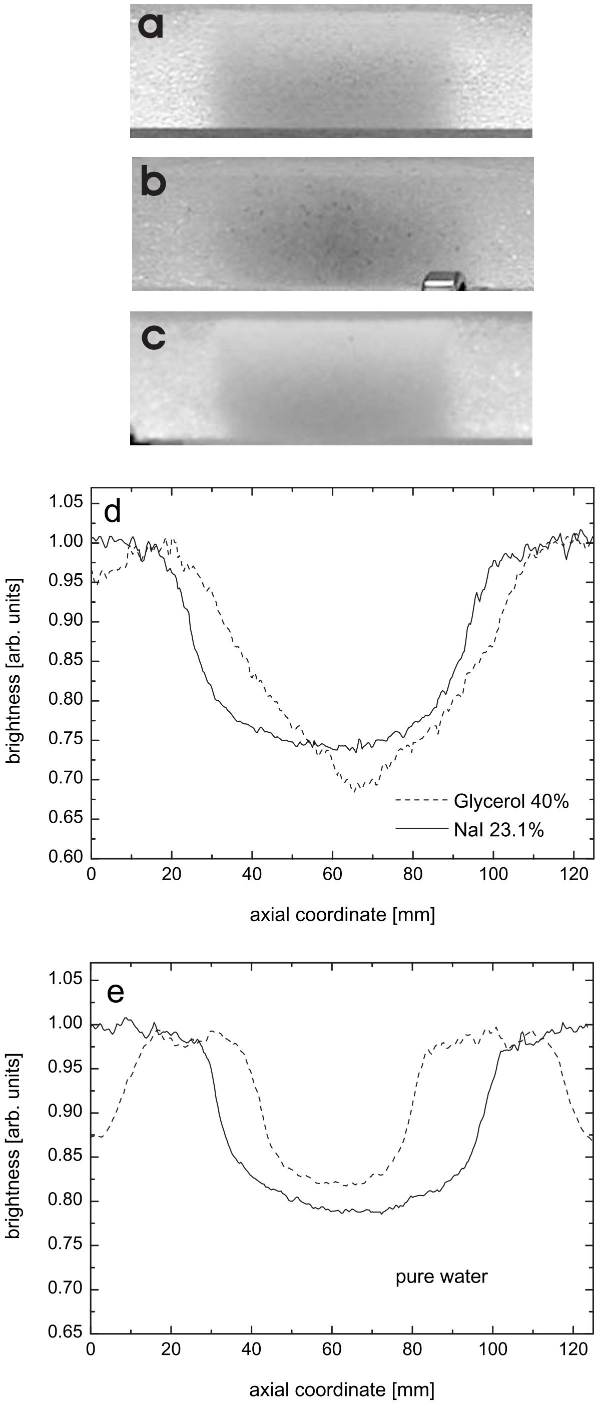

FIG. 10. Segregated bands of small particles as seen in selected sections of the tube. (a) 23.1% NaI/water solution after 11 000 rotations at 20 rpm, second stripe from the left-hand side in Fig. 8. (b) 40% glycerol-water mixture after 450 000 rotations at 30 rpm, left-hand stripe in Fig. 7. (c) Pure water system at 15 rpm after 16 000 rotations. Image widths are 123 mm. Note the sharp boundaries between typical bands in the low viscosity fluids (a) and (c), in contrast to the gradient in the band composition in the high viscosity mixture (b) even after much longer rotation time. (d) Optical intensity profiles taken along the central section of the tube, averaged over a vertical range of 12 mm. The intensities have been scaled to the same maximum. (e) Optical profile of the water system after 6700 rotations (dashed line) and 16 000 rotations (solid line).

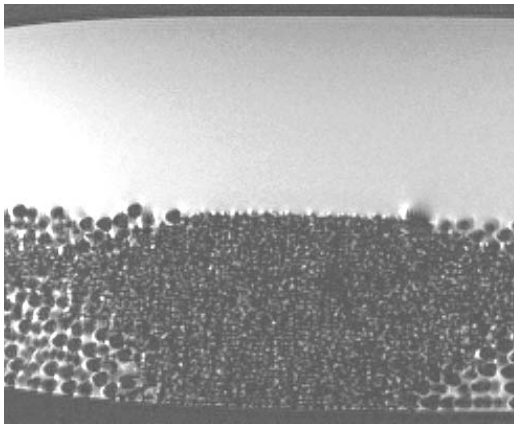

FIG. 11. Magnetic resonance image of a stripe of small beads in the pure water system, vertical slice in the tube midplane. Image size approximately 45 mm × 37 mm. Details of the experiment have been described in [25].

NaI solution agrees with the pure water system. In the same way as for the pure water system, rotation at velocities above 25 rpm leads to a certain stabilization of uniform stripe arrays. The pattern usually starts to decompose one by one stripe in sequence, not by random dissolution, as seen, for example, in Fig. 9.

For the glycerol systems, the increased viscosities lead to a slower decay of the band structure at progressed times, which is evident even in the logarithmic plots. These deviations in the long term limit evidence that the higher viscosity leads to a certain stabilization of the band pattern against coarsening processes.

IV. CONCLUSIONS AND SUMMARY

The experiments performed with liquids of different viscosities and densities have shown that the change of the fluid *density* by about 20% as well as the addition of salts in large

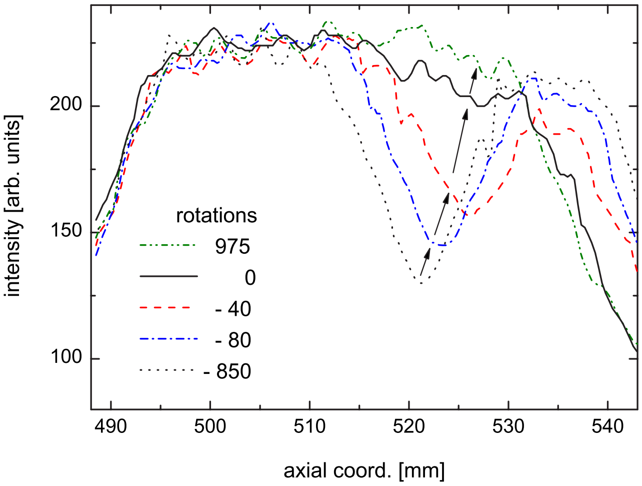

FIG. 12. (Color online) Profiles before and after the vanishing of a single stripe (the stripe at axial position 52 cm in Fig. 5). The figures denote numbers of rotations before and after the first image where the stripe has disappeared.

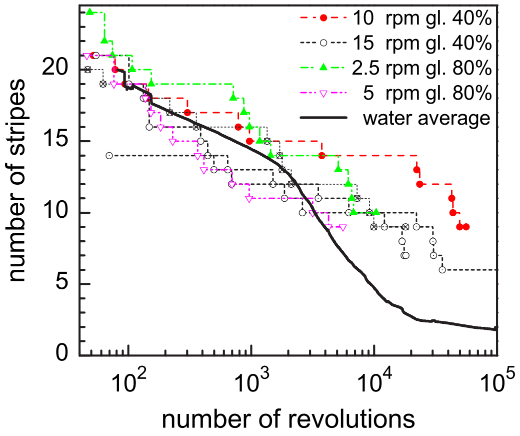

FIG. 13. (Color online) Coarsening of the granular stripe pattern in a 662 mm long drum filled with different water-glycerol mixtures. The thick solid line represents an average of all measurements performed in the pure water system (at 10 to 20 rpm), the other lines represent individual experiments for different concentrations of glycerol and different rotation speeds.

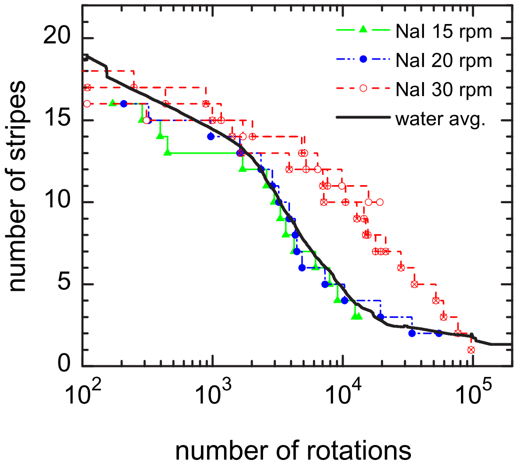

FIG. 14. (Color online) Coarsening of the stripe pattern in a 662 mm long drum filled with NaI solution in water. The thick solid line represents an average of all measurements performed in the pure water system (at 10 to 20 rpm), the other lines represent individual experiments for different rotation speeds.

quantities does not influence the formation and evolution of segregation patterns of the granulate in the rotating drum experiment qualitatively. There are some small quantitative changes, well within the scattering of data for repeated individual measurements of a given system and parameter set. No trend is indicated for solutions of different densities in the investigated range.

On the other hand, the variation of the *viscosity* of the liquid plays a crucial role particularly in the pattern dynamics, but also for the structure of the segregation patterns. The initial (≈1000 rotations) coarsening dynamics is only slightly influenced on the scale of drum rotations, but there is a considerable shift of the dynamic range (in terms of rotation speed of the drum) within which the segregation patterns are observed. This shift is of the same order of magnitude as the change in viscosities. An order of magnitude discussion shows that in the 80% glycerol system with a viscosity increased by a factor of 20 respective to water, the dynamic range is shifted by almost one order of magnitude. This may serve as a rough estimate when one compares different liquids, but certainly the result cannot be extrapolated very far. Definitely it cannot be applied to compare slurries and dry granulate, although the trend of accelerated dynamics with decreasing viscosity of the surrounding fluid is maintained. Additionally, the viscosity of the liquid tends to stabilize the band structure in its long-term evolution.

Whereas the overall coarsening dynamics of the bands is not qualitatively altered in the statistical average, the spatial structure as well as the temporal evolution of a dissolving stripe are qualitatively influenced by the viscosity of the interstitial liquid: With increasing viscosity and increasing rotation speed of the container, the borders between the segregation bands in the glycerol mixtures have a tendency to become smoother. In particular in the fast spun (30 rpm) glycerol mix, the segregation process is far from being complete. The fact that the viscous properties of the liquid have comparably small influence on the velocity of pattern formation and coarsening measured in units of drum rotations, but alter the internal band profiles and efficiency of the segregation is somewhat counterintuitive and must be analyzed further by proper microscopic models.

The effect of the sudden collapse of segregation bands after long periods of slow gradual thinning also has not yet been addressed experimentally or theoretically. From our experimental data it becomes evident that the small size beads in a collapsing band are moved, quasicollectively, towards the nearest neighboring band (or distributed between the two neighboring bands) of small beads. After a few dozen rotations, the remainder of the band is adsorbed completely by its neighbor(s). This effect is more or less pronounced in all slurries investigated in our study, but most pronounced in the viscous glycerol mixtures. More experiments, including Magnetic Resonance Imaging, must be performed in order to characterize details of these decomposition processes.

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