Self-Organization in Granular Slurries

Nitin Jain,[1] D. V. Khakhar,[1,*] R. M. Lueptow,[2] and J. M. Ottino[1,2,†]

[1]*Department of Chemical Engineering, Northwestern University, Evanston, Illinois 60208*
[2]*Department of Mechanical Engineering, Northwestern University, Evanston, Illinois 60208*

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We report the existence of self-organization in wet granular media or slurries, mixtures of particles of different sizes dispersed in a lower density liquid. As in the case of dry granular mixtures, axial banding (alternating bands rich in small and large particles in a long rotating cylinder) and radial segregation (in quasi-2D containers) are observed in slurries. However, when compared with the dry counterpart axial segregation is significantly faster and the spectrum of outcomes is richer. Moreover, experiments with suitable fluids reveal, for the first time, the internal structure of axially segregated systems, something that up to now has been accessible only via magnetic resonance imaging experimentation.

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Slurries, in which particles are dispersed in a liquid, are of considerable importance. Technological examples appear in cement, ceramics, fine chemicals, hydrometallurgy, treatment of hazardous materials, and in the food industry [1]; examples in nature appear in evolution of landslides and transport in river sediments [2]. Mixtures of tumbled granular materials under flow exhibit various intriguing types of unmixing or self-organization. Small differences in particles' density, size, surface characteristics, or shape may trigger the effect [3,4].

There have been several studies addressing the case of segregation in dry systems and understanding is beginning to emerge [5]. The prototypical systems consist of spherical particles of different sizes (S systems, diameter ratio r_S) or different densities (D systems, density ratio r_D) in 3D or quasi-2D rotating cylindrical containers. Upon flow particles radially segregate in the cross section (radial segregation) of the container and separate in axial bands along the axis of the container (axial segregation). Quasi-2D experiments and theory have been reported for both S and D systems [6,7]; however, all axial segregation experiments correspond to S systems [8]. The wet case is unexplored. In fact, the only closely related study pertains to the case of monodisperse dilute neutrally buoyant suspensions of equal size particles in a partially filled horizontal Couette flow [9]. The physics of this case, however, is entirely different since gravitational sedimentation is very important in our system, and our system has no gas-liquid interface which appears to be essential for segregation in the case of Ref. [9].

The bulk of the experiments reported here correspond to binary S systems with $r_S = 0.2$. The particles are monodisperse glass beads (1 mm diameter) and mixtures of glass beads: 1 mm (transparent) and 0.2 mm (colored). Equal volumes of the large and small beads are used in all the mixtures. Three systems are considered: dry, where the fluid is air, and two wet systems, with the fluids being water and a 67% by weight solution of sodium iodine (NaI) dissolved in water to match the refractive index of the transparent glass beads. This enables visualization of the internal structures formed. In all the experiments the packed bed of solids occupies 50% per volume of the cylinder, with the liquid filling the entire volume of the cylinder.

Two types of gravity driven flows generated by rotation, with the cylinder axis horizontal, are considered: 3D flow in a long cylinder for axial pattern formation experiments and quasi-2D flow in a short cylinder for transverse flow and radial segregation experiments. The 3D system comprises a transparent Perspex cylinder of diameter 5 cm and length 40 cm; the quasi-2D system a cylinder of diameter 28 cm and thickness 7 mm with a transparent Perspex faceplate. The rotational speed of the cylinders is controlled using a stepper motor (Compumotor).

In the 3D experiments the cylinder is rotated at a high speed (2.1 rps for water and 0.53 rps for the NaI solution) to achieve an initial state that is axially uniform. The cylinder is then rotated at a much lower rotational speed and the dynamics of the formation of bands is studied by digital photography of the flat free surface and image analysis. The image analysis involves subtracting the background (the image taken just after the cylinder starts rotating) from an image, and from this computing the average intensity profile along the axis of the cylinder, with the average taken along the diameter of the cylinder. The standard deviation of the intensity from the mean, σ, called the intensity of segregation [10], is then calculated as a measure of the extent of segregation. Slurries with water and NaI solution at different speeds of rotation are used. A similar procedure is used for the dry beads, but in this case the initial uniform state is achieved by rotating at a sufficiently low speed such that no bands are formed. In the experiments with NaI, once bands are formed, rotation is stopped and the cylinder is immersed in a rectangular glass walled tank of water (to reduce the distortion by the curved surface of the cylinder) and photographed using different lighting conditions as indicated in Fig. 1. The speeds in all cases are selected so as to be in the cascading regime. They are lower for the NaI solution compared to water because both density and viscosity of the NaI solution are higher than those for water.

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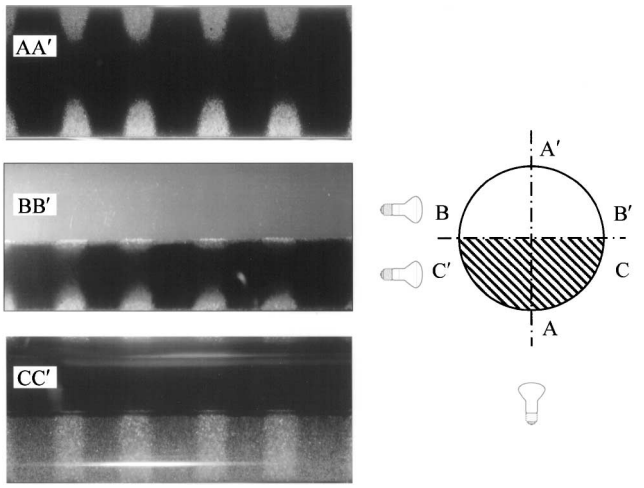

FIG. 1. Self-organization in a slurry consisting of equal parts per volume of 0.2 and 1 mm glass beads. The liquid, used to match the refractive index of the clear beads, is a 66% by weight solution of NaI in water. Pictures are taken from the primed position, and the direction of lighting is indicated by the light bulb. (AA′) Top view and (BB′) side view show the internal structure consisting of a core of small beads within clear big beads. (CC′) Side view shows only the surface view.

The 2D experiments are used to obtain the streak lines for the transverse flow, using time lapse photography, and to investigate radial segregation. The radial segregation experiments involve mixtures of small and large glass beads under dry and wet conditions.

The 3D experiments reveal that slurries containing a mixture of particles rapidly self-organize to form a pattern that appears on the surface as alternating bands of the small and large particles (Fig. 1, CC′). Visualization of the structure using refractive index matching and backlighting reveals a central varicose core of the smaller particles with rings of the larger particles (Fig. 1, AA′ and BB′). The structure is reminiscent of that found for dry systems using magnetic resonance imaging [8,11].

The dynamics of band formation for the wet (water) and the dry systems are shown in terms of the variation of intensity of segregation (σ) with number of cylinder revolutions (N) in Fig. 2. In both systems the rate of segregation increases with rotational speed, as does the fractional surface area (a) covered by the bands of the smaller particles (see Fig. 1, CC′). There are, however, notable differences: segregation occurs at a higher rate in the wet system (a factor of about 2 when compared on a revolution basis), and the fractional area coverage is always greater in the wet system. The dashed lines in Fig. 2 are predictions of the intensity of segregation assuming complete segregation, given by $\sigma_\infty = [a(1 - a)]^{1/2}$, where a, the surface area occupied by the smaller particles, is measured from the final images. There is good agreement between prediction and experimental values. Results for wet systems with NaI solution are similar to those for water but for an overshoot in the intensity of segregation versus N curve. This results from the formation of initially narrow bands which

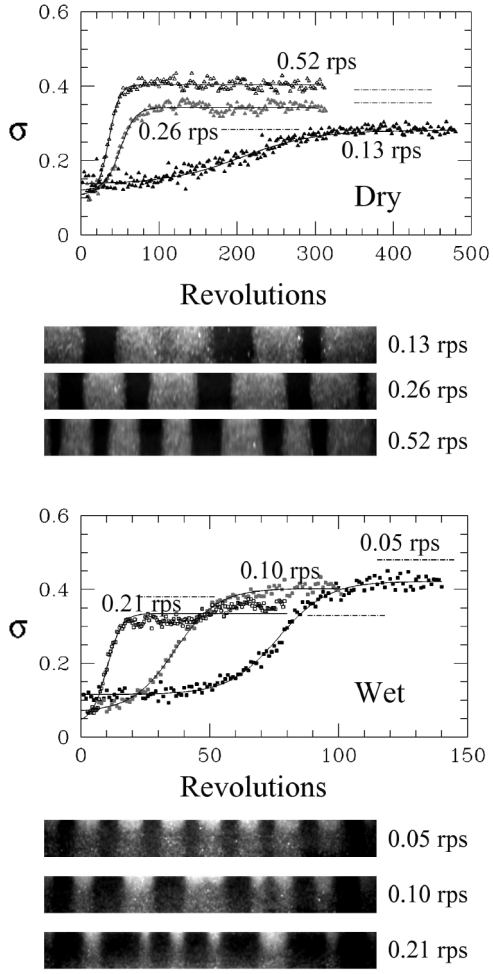

FIG. 2. Dynamics of axial banding for dry (air) and wet (water) systems. The solid lines are fits of Eq. (1) given below to experimental data (symbols). The dashed lines are the predicted values assuming complete segregation based on the final area coverage of the top surface (a) by the small beads (calculated after thresholding the pictures). Final segregation patterns (images below the plots) at different angular velocities for both systems show an increase in the area coverage of bands with rotational speed. Note the different scales on the revolution axis.

broaden with time, thus a increases and σ_∞ decreases after reaching a maximum which corresponds to the formation of the narrow bands. The solid sigmoidal curves in Fig. 2 are fitted curves of the form

$$\sigma = \sigma_0 + \Delta\sigma \tanh[k(N - N_c)], \qquad (1)$$

where k, σ_0, $\Delta\sigma$, and N_c are fitting parameters. The data are well described by Eq. (1), and the rate segregation given by the parameter k obtained by fitting is shown in Fig. 3 versus the measured area coverage. The rate of segregation is higher for the wet systems, particularly for the NaI solution, and the area coverage for the wet systems is high, with $a \geq 0.5$ in most cases. Since the small particles comprise only 50% by volume of the mixture and a central core of the small particles runs through the large particle bands, we are led to the conclusion that a fraction

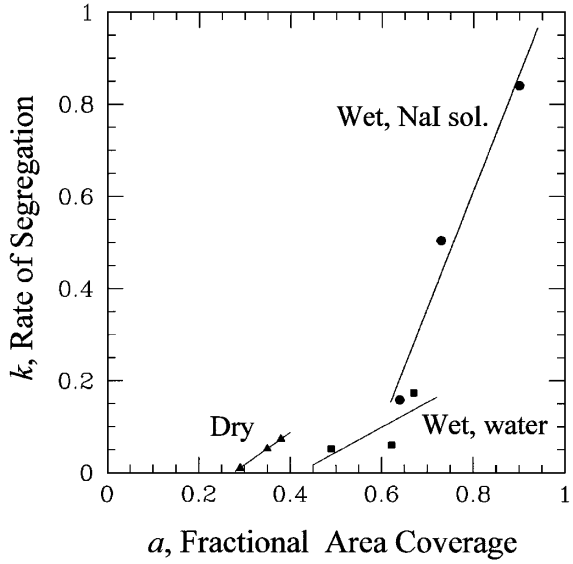

FIG. 3. Plot of rate of segregation (k) versus the final area coverage (a). The data points correspond to different angular velocities for dry (△) and wet [water (■) and NaI solution (•)] systems. Final area coverage (a) is always below 50% for the dry material but can be as high as 90% for the slurry.

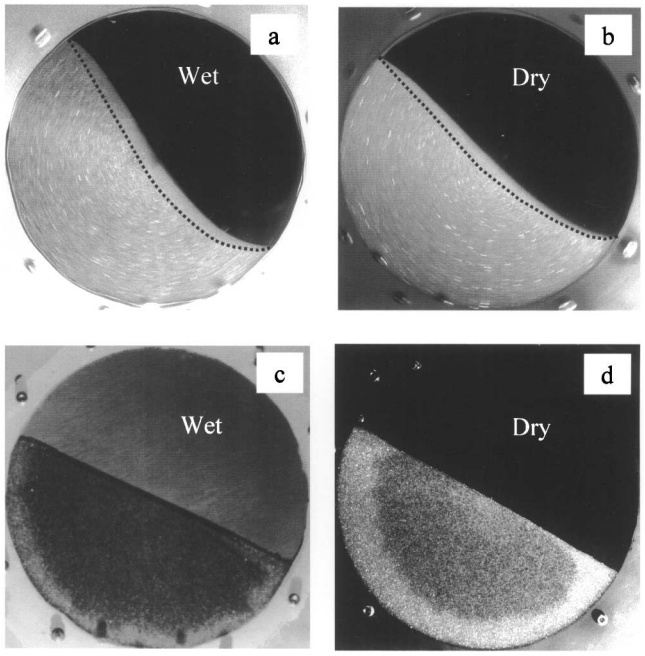

FIG. 4. Quasi-2D tumbling experiments. (a),(b) Streak lines from time lapse photography; 1 mm glass beads are rotated at 0.04 rps. The dashed line shows the interface between the flowing layer and the bed. The wet system has a thicker flowing layer (by a factor of 1.4). The angular speed corresponds to the middle value used in the experiments for the wet system shown in Fig. 2, assuming a constancy of Froude number. (c),(d) Radial segregation for 0.2 and 1 mm glass beads rotated at 0.008 rps. The angular velocity is selected to have a flat, continuous flowing surface for both wet and dry beads.

of the larger particles must lie within the bands of the smaller particles.

The internal structure of the bands was studied by freezing. Water was first allowed to slowly drain out by loosening one of the end caps, then hot gelatin solution was injected to submerge the particle bed, and the cylinder was placed in a freezer. Upon slicing and separating the particles by sieving, we found a significant fraction of large particles (33 vol % for water at 0.21 rps) uniformly distributed in the bands of the smaller particles. Thus the large particle bands are pure with a core of small particles, while the small particle bands are a mixed phase rich in the small particles (a similar result has been reported for dry systems [8]). The fractional area coverage (a) is thus indicative of the extent of mixing of the large particles in the small particle bands, and experiments indicate that such mixing is greater for slurries, and increases with rotational speed.

The flow and segregation in the cross section of a cylinder using wet and dry systems in the quasi-2D cylinder is shown in Fig. 4. Figure 4a shows the streak lines for the transverse flow. A thin surface flow with solid body rotation is discerned from the photograph. This is qualitatively similar to the dry system (Fig. 4b), but the layer thickness is significantly greater (by a factor of about 1.4) in the wet system at the same rotational speed (0.04 rps). The slurry with a mixture of two sized particles radially segregates with the small particles forming a radial core (Fig. 4c), as does the dry system (Fig. 4d). The core is larger for the wet system for the same rotational speed (0.008 rps), indicating a greater degree of intermixing of the large and small particles in this case. This is consistent with the higher intermixing found for wet systems in the axial segregation experiments.

The above results show that all behaviors observed for dry systems carry over (with some differences in detail) to the case of wet systems. The Bagnold numbers [12,13] for the wet ($\mathrm{Ba} = 10^{-2}$) and dry ($\mathrm{Ba} = 10^{2}$) systems are very different, suggesting that viscous stresses dominate in the wet case and collisional stresses in the dry case. However, in both of the systems considered here—systems where the motion is induced by tumbling and flows are relatively slow—gravity plays a significant role and particles, as they roll past each other, may be in contact with each other. This is true even for wet systems, where lubrication stresses are of the same order of magnitude as gravitational stresses ($\tau_g = \rho_p g d$) in the flowing layer. In the fixed bed, the weight of the particles forces out the fluid between particles until (rough) surfaces touch and frictional forces dominate. Thus the kinematics of the wet and dry systems is similar but very different from partially wet systems where capillary forces result in aggregation of particles.

A precise theoretical analysis appears difficult. An alternative route is to estimate the order of magnitude of mixing fluxes, due to diffusion, and segregation fluxes, due to differential body forces. A scaling analysis (given below) shows that in wet and dry particulate systems these ratios

are similar in magnitude, but vastly different from molecular and colloidal systems.

Consider first the diffusion fluxes. The thermal motion of molecules results in mixing characterized by a diffusivity, D_{AB} ($\sim 10^{-5}$ cm^2/s for normal liquids) and in the case of Brownian particles ($d < 1\mu$m) by the Stokes-Einstein relation, $D_{\mathrm{Br}} = k_B T/3\pi\mu d$, where k_B is the Boltzmann constant and T is the absolute temperature. Particles in non-Brownian particulate systems (as considered here) are static since thermal motion is negligible ($kT/\rho_p g d^4 \sim 10^{-20}$). In flowing particulate systems, however, interaction between particles (mediated by collisions or viscous forces) causes random motion that is analogous to thermal motion, characterized by a diffusivity $D_{\mathrm{coll}} \sim \dot{\gamma} d^2$ for both wet [14] and dry [15] systems. The diffusion velocity for each of the three cases is given by $v_{\mathrm{diff}} \sim D/d$, where D is the appropriate diffusivity.

The diffusion velocity, causing homogenization, should be contrasted with the velocity of the process trying to induce inhomogeneity due to size differences. Considering dilute systems (low particle volume fractions) for which analytical expressions are available, the relative segregation velocity for molecules is [16] $v_{\mathrm{seg},AB} \sim D_{AB} m g(1 - r_S^3)/k_B T$, where m is the mass of a molecule. Assuming Stokes' flow for the Brownian particles, the relative segregation velocity for the particles is $v_{\mathrm{seg,Br}} \sim (\rho_p - \rho) d^2 g(1 - r_S^2)/\mu$. For dry granular shear flows we have [17,18] $v_{\mathrm{seg,coll}} \sim g(1 - r_S^3)/\dot{\gamma}^2 d$; a similar expression is not available for wet systems. In dilute systems, since the motion of particles is unhindered by other particles, kinematics in the wet and dry systems should be very similar. However, since the segregation is driven by gravity, buoyancy forces would be important. Here we take the segregation flux for the wet system to be $v_{\mathrm{seg,coll}} \sim g(1 - \rho/\rho_p)(1 - r_S^3)/\dot{\gamma}^2 d$, which is the same as the dry system but accounting for the lower body force felt by the particles in the wet system due to buoyancy. The segregation number defined as $\mathrm{Se} = v_{\mathrm{seg}}/v_{\mathrm{diff}}$ is a measure of the tendency of a system to segregate (or the inability of diffusion to erase fluctuations). Thus, in general, molecular systems ($\mathrm{Se} \sim 10^{-13}$) never segregate due to gravity, Brownian systems have a moderate tendency to segregate ($\mathrm{Se} \sim 3$), but both dry ($\mathrm{Se} \sim 20$) and wet ($\mathrm{Se} \sim 90$) particulate systems, as used in this work, are highly prone to segregation. Though this was not studied in detail, the mechanisms by which axial segregation occurs appear to be similar in the wet and dry systems: small axial flows produced by undulations of the flowing particle surface.

Experimental results reveal that axial segregation in slurries is robust. The phenomenon is significantly faster than in dry systems. The effect of rotation is more revealing—significant changes in the thickness of the bands serves to underscore that significant changes in the interior have to accompany the process. Experiments with NaI demonstrate details of the structure that up to now have been available only via sophisticated experimentation. Freezing experiments show that bands comprise a mixture of small and large particles, the concentration of the large particles is high for wet systems relative to the dry case, and the concentration increases with rotational speed. There are several possible extensions of this work. An important one is to focus on the competition between chaos and self-organization as has been reported for dry systems in non-circular containers [19].

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*Permanent address: Department of Chemical Engineering, Indian Institute of Technology–Bombay, Powai, Mumbai 400076, India.
†Corresponding author.
Email address: jm-ottino@northwestern.edu

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