INTRODUCTION
Rich physics of static/dynamic behavior of granular materials have promoted a great interest among researchers in different fields. Most recently, compaction, segregation and a various types of pattern formation have captured attention of many physicists. Segregation seems to exist anytime when different species of particles are in the relative motion. Bulk material handling plants frequently experience non-uniform products due to size, shape, and density variations among particles.

A counter-intuitive axial segregation phenomenon in a rotating horizontal cylinder has recently been under an intense scrutiny by many researchers in different disciplines. However, the first detailed observation of this phenomena was made by Oyama in 1930. He conducted a series of experiments using a short cylinder made of cast iron with its diameter and length of 200mm and 400mm, respectively. After 15 minutes of rotation, he recorded patterns of axially segregated bands created by the mixtures of different sizes of limestones, different weight ratios (6.14, 2.59 and 2.37), different filling ratios (4kg, 6kg, and 8kg) and rotating speed (10, 20, 40, 60 and 100rpm).

Donald and Roseman, Bridgewater, and Roger and Clement extended Oyama's work and investigated mechanisms of the axial band segregation. Donald and Roseman concluded that the axial banding only occurred if the static angle of repose was larger for the smaller particles than that for the larger particles. Bridgewater expanded the explanation given by Oyama and proposed a mechanism to allow smaller particles to accumulate near the end cap. Extra friction again the end caps brings particles right next to the caps to a higher level, and the larger particles tend to roll down the slope easier than the smaller particles do. These two effects produce a high concentration of smaller particles next to the end caps. Along the cylinder axis, statistical variations in the concentration were proposed to cause the axial band formation for the similar reason described above. Roger and Clement also observed axial segregation but the cylinder they used was probably too short compared to the length to isolate the effects due to the boundary walls. Das Gupta et al. further expanded the argument by the previous authors and actually measured the dynamic angle of repose of a single component of different size of particles. They found that the dynamic angle of repose depended on the rotation speeds and it did not differ much when it was smaller than a critical value. However, when the flow was driven harder to cause the higher dynamic angle of repose, then the smaller particles started to show the higher the dynamic angle of repose.

Savage reported that axial segregation occurred for the 50-50 volume mixture of spherical and rod-shaped particles of similar size. Based on the visual observation that rod-shaped particles always formed axial bands adjacent to the two end caps where the dynamic angle of repose was higher suggesting more resistance of the rod-shaped particles to rolling. He formulated a diffusion like equation which contains both a Fickian diffusion flux term and a preferential drift term. When the drift due to angle of repose exceeds the diffusion flux, then the combined terms described above gives a negative effective overall diffusion coefficient which promotes the axial segregation.

There is a growing consensus that the interplay between the particle dynamics and the evolving internal structures during the segregation process must be carefully investigated. Magnetic resonance imaging (MRI) has recently been used to non-invasively obtain much needed static/dynamic information such as concentration, velocity and fluctuations in velocity. It has proven to be capable of depicting the evolution of segregation processes in a rotating cylinder. Segregation in a straight horizontal rotating cylinder involves two processes: the first is to transport small particles in the radial direction to form a radial core, and the second is to transform the radial core into axially segregated bands. Percolation and/or “stopping” have been suggested as possible mechanisms for the radial segregation. As to driving mechanisms for axial band formation, however, much less is known. It has been proposed that the dynamic angle of repose promotes this process, and Hill and Kakalios have reported that particles mix or demix depending upon the competition between angle of repose. We claim that the dynamic angle of repose could be one of the causes for a particular range of solid fraction and rotation rates, however, it fails to offer reasonable explanations for certain phenomena associated with the axial migration. For example, we always observe that the radial segregation preceds the axial segregation. The radially segregated core of small particles then transforms into axially segregated bands. By definition, the effects of the dynamic angle of repose is restricted near the free surface where the flowing layer is present. However, the process of transformation from the radially segregated core to the axially segregated bands occurs by migration of small particles located in the deep radial core region.
We have designed a series of experiments so that the effects of the dynamic angle of repose can be localized in a very confined region by filling the cylinder almost completely full. Under these extreme conditions, small particles still form a radial core and also migrated to form axial bands. This can not be explained simply by the argument based on the dynamic angle of repose. We present our recent non-invasive experimental images to show a new way of forming axial bands.

As to 2D experiments we have identified in our experiments that particles located near the center of the cylinder axis behave quite differently from the ones located near the boundary. The particles near the center seem to move slower and randomly. On the contrary, the particles located far from the center are engaged in the rapid shearing motion.

**EXPERIMENTAL**

We have first investigated behavior of granular particles in a long horizontal cylinder. We prepare initial samples whose volume is almost completely occupied by the particles. The cylinder of length of 27cm with an inner diameter of 7cm is made of acrylic material. To achieve a maximum initial packing so that we can minimize the size of the flowing layer, pre-mixed particles are poured into the cylinder. Near consistent tappings on the side of cylinder during the pouring provide a desired packing. Once the initial sample is prepared in this way, it is placed on a pair of rollers which were placed on the horizontal table and rotated at 20 rpm.

Particles used for this experiment are poppy seeds, mustard seeds and pharmaceutical pills which give excellent NMR signals. Poppy seeds are flat, angular and smallest with about 1mm of effective diameter. Mustard seeds are relatively round and have about 1.7 mm diameter and a bulk density of 1.3 g/cm³. Pharmaceutical pills of 1 and 4 mm are used for the detailed investigation in conjunction with MRI presented in this article. The 4mm particles are painted black by a commercially available permanent oil based ink to help visualize for non-MRI experiments. There is no signs of alternation of the surface properties after these particles are painted. The pills contain a liquid core of medium chain triglyceride and the gelatin outer shell weighs about 30% of the total weight of a particle of either size. Recently, Louge et al. 15 conducted a detailed binary impact experiment using these particles and estimated the normal coefficient of restitution to be around 0.89. When the cylinder is partially filled and the dynamic angle of repose seems to have significant effects on segregation processes, material properties of particles such as the coefficient of restitution and friction were important factors. In the current investigation, however, it is less known that which properties play more important roles.

Nakagawa et al. 16 first conducted non-invasive MRI of flows of mustard seeds in a horizontal rotating cylinder. MRI was also used to study shaking of granular materials. 17 Later, a similar MRI was applied first by Nakagawa et al. 6 and later by Hill and Kakalios9 to investigate evolution of radial and axial segregation by studying internal structures during the process. A similar technique was used in this experiment. Recently, Ovryn et al 18 have proposed to develop a versatile, user-friendly and inexpensive NMR machine to promote research in Microgravity Fluid Physics discipline.

For 2D experiments, we use a very short cylinder of the gap and diameter of 5 mm and 11 cm, respectively. Pharmaceutical pills of 2mm are used. We painted some of these particles with the permanent black ink as described earlier to place a line of these painted particles going through the center of the cylinder. This line is placed to study the motion of particles located near and far from the center of the cylinder. To follow the motion of particles we use a high resolution 8mm camcorder.

**RESULTS AND DISCUSSIONS**

One of the main purposes of conducting a series of segregation experiments with the proposed high filling ratios is to gain more insights for the possible existence of a mechanism which is capable of producing radial and axial segregation phenomena independent of the effects of the dynamic angle of repose. Regardless the filling ratio, the cylinder rotation drives particle migration. In the event of the moderate filling ratios of about 50% as have been investigated extensively, there exists a substantial flowing layer which induces particles migration in both radial and axial directions. As indicated earlier by Nakagawa et al. 16 the flow depth increases rapidly as the rotation speed increases slightly. However, as soon as the flow is developed, the dilated flow becomes deep enough to carry small particles into the core to form a radial core.

On the contrary, in an almost completely filled cylinder there is very little room for the surface flow to develop. Initially, when the cylinder starts rotating, sparsely located small particles accumulate near the bottom of the very minute flowing layer to form streaks of different lengths. After a few rotations, a series of streaks merge to form a thin ring structure. When this is completed, there is hardly any small particles outside the ring since all the small particles have migrated in the radial direction to form the ring.

In the region outside the ring, there is hardly any small particles visible to the eyes at the ends. Inside the ring, there are still some small particles present since they did not participate in the active migratory mechanism as described earlier. The concentration of
small particles in this region, however, increases in time. It appears that the small particles are accumulated in the core through axial migratory motion. The radial ring formation and axial-filling of the core are observed at both ends of the cylinder. With this high filling ratio, the axial-filling is truly three-dimensional mechanism and has not previously been observed by any 2D rotating cylinder experiments. The radial migratory behavior of small particles in a short 2D drum may impose severe restrictions on particle drift in the axial direction. It is conceivable that even for the case with moderate filling ratios, the axial-filling could take place in the formation of the radial core. So, it might be more realistic to view that even the radial core formation process must be discussed in conjunction with the axial migration of particles as a true 3D phenomenon.

When the initial axial filling is completed at both ends, the radial core at one end starts disappearing faster than the other, indicating axial migration of small particles. The end result is shown in Fig.1 below. Well separated bands of large and small particles are formed together with a rather sharp interface region where there is a core of small particles still present. Unlike the final configuration obtained for a partially filled cylinder experiments with the odd number of bands, we have almost completely separated two bands. Since there is no surface flow influenced by the friction of the end caps, there is no reason to observe a larger concentration of smaller particles right next to the end caps. However, based on the symmetry argument, this two band configuration does not seem to be as stable as three bands. What we reported here may still be an intermediate stage of an evolving process.

![Figure 1 NMR image of axial band segregation in a nearly packed cylinder.](image)

Figure 1 NMR image of axial band segregation in a nearly packed cylinder.

Figure 2 shows a series of line configurations at different times of the dispersion process. The initial straight line (Fig. 2a) soon becomes shorter through the dispersion of particles near the boundary. The shortened line then shifts downward and rotates in the direction of the cylinder (Fig. 2b). This line continues to become shorter and move around in the central region where the fast shearing is not affecting its motion (Figs. 2c and 2d), and eventually disperses to the surrounding particles to form a mixture.

![Figure 2. Motion of particles in a short cylinder.](image)

Figure 2. Motion of particles in a short cylinder.

In conclusion, a brief description of a close up observation of particle motion is given. With this high filling ratio, there is not enough room for particles to establish a surface flow, however, using little space or voids available for them they constantly rearrange themselves through a series of collapses of local structures (micro-collapses). It is observed that these micro-collapses occur everywhere in the cylinder perimeter. Keeping the same high filling ratio, we have also conducted a series of experiments where the two axially segregated initial ends transformed into a radially segregated core. A close look at the interface between the bands of large end small particles in the beginning also provides a very clear picture as to how each species diffuses through the voids created by a series of micro-collapses. This diffusive motion of particles in the central region was also observed in the 2D experiment.

ACKNOWLEDGEMENTS

MN and JLM were supported, in part, by NASA through contract NAG3-1970. MN would like to acknowledge members of the Particulate Science and Technology Group of Colorado School of Mines, in particular, Jon Eggert and David Wu for their critical discussions. We would also like to acknowledge Taiho Pharmaceutical Co. for their generous donation of pharmaceutical particles.

REFERENCES