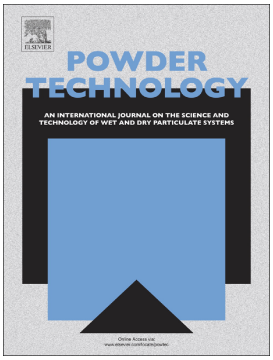

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PII: S0032-5910(18)30228-6
DOI: doi:10.1016/j.powtec.2018.03.035
Reference: PTEC 13266

To appear in: *Powder Technology*

Received date: 2 July 2017
Revised date: 5 January 2018
Accepted date: 18 March 2018

Please cite this article as: Humair Nadeem, Theodore J. Heindel , Review of noninvasive methods to characterize granular mixing. The address for the corresponding author was captured as affiliation for all authors. Please check if appropriate. Ptec(2017), doi:10.1016/j.powtec.2018.03.035

Review of Noninvasive Methods to Characterize Granular Mixing

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Keywords
Granular flow, Particle-particle mixing, Spectroscopy, Tomography, Velocimetry

Abstract

Granular mixing is a common process observed in a variety of industries such as construction, chemical processing, food handling, and cosmetics and pharmaceutical manufacturing. Over the years several methods have been devised to characterize granular mixing and to provide information on the quality of the mixture as well as mixing end points. In this work, three different noninvasive measurement categories are reviewed, namely: velocimetric, spectroscopic, and tomographic techniques. Velocimetric techniques such as Particle Image Velocimetry (PIV) and Radioactive Particles Tracking (RPT) are able to provide the trajectories and velocities of individual particles during the mixing process. Spectroscopic techniques focus on the chemical composition of the sample and are well suited for applications where there is little difference in the physical properties of the constituents such as particle size and density. Tomographic techniques such as X-ray Computed Tomography (CT) and Magnetic Resonance Imaging (MRI) provide information such as the spatial distribution of the different constituents in the sample and are useful in determining dead zones and inhomogeneities. A few other techniques are also discussed, particularly passive acoustic methods which, though can neither provide spatial distribution nor individual particle trajectories, are still useful and can be employed to determine mixing end points. The objective of the work is to provide a comparative assessment of the various noninvasive techniques and discuss their ability to effectively characterize the mixing process. The reported techniques will be reviewed thoroughly based on their functionality, viability, and characterization capability. The advantages and disadvantages of the

various techniques are summarized and a comparison of the utility of the techniques in various applications is discussed.

INTRODUCTION

Granular mixing has been a part of human life since our earliest ancestors put pestle to mortar. In recent years, granular mixing has gained importance, particularly in the highly-regulated industries of food and pharmaceuticals. The study of granular mixing is therefore crucial when it comes to quality control of items from breakfast cereal to medicine tablets. Other industries where granular mixing plays an important role include construction (blending of building material), cosmetics (manufacture of makeup products), and household hygiene items (laundry detergents and drain decloggers) [1]. In most cases the focus is on ensuring the homogeneity of the manufactured product. Since the major causes of incomplete or inefficient mixing include inefficient mixer design and segregation during the mixing process [2], particle pathlines and velocity profiles in the mixer vessel also need to be scrutinized.

Intuitively, it made sense to check for mixing homogeneity by extracting samples from the vessel. However, over time different issues with sampling methods such as destruction of the bed on sampler insertion, and with invasive techniques in general such as disturbance of the granular flow path, paved the way for the development of noninvasive techniques [3]. Noninvasive methods have recently become more popular as many of them can be deployed in-line (i.e., the mixer does not need to be stopped for data acquisition). Coupled with the fact that these techniques can be used in various kinds of mixing equipment makes them a better option.

This work reviews noninvasive techniques available today to assess mixing in industrial processes. Our goal is to compare the advantages and limitations of each of the described methods and provide some direction as to which method may work for a particular need or application.

VELOCIMETRIC TECHNIQUES

All velocimetric techniques reviewed below involve methods used to identify a limited number of particles and track their movement through a mixer as a function of time. Velocimetric methods can

be broadly divided into two categories. The first category consists of methods in which electromagnetic energy is added to the system, and their interaction is recorded. The electromagnetic radiation could either be visible light, X-rays or even lasers. The second category involves energizing individual particles and then recording how the particle energy emission decays over time. This category is then further divided based on the type and process of the radiation emission and includes radioactive decaying particles and positron emission particle tracking.

Optical Image Analysis

Earliest noninvasive techniques focused on the use of cameras to obtain videos and photographic stills of the mixing phenomenon. The vessel being studied usually had a transparent wall or a free surface to allow for image acquisition as shown in Figure 1. Experiments consisting of multi-color granular material are typically used to quantify mixture homogeneity; while experiments with a small number of detectable "tracer particles" are employed to study the trajectory of particles in the vessel.

Horizontal axis bladed mixers were studied by Rumpf and Mueller [4] in the 1960s. The vessel was made of transparent Plexiglas and observations were taken from a side wall. Several types of blades were used to study the movement of the particles during blade rotation. Similar studies by Malhotra et al. [5,6] added colored tracer particles to the bulk medium. The colored particles were then used to trace the particle flow patterns and velocity profiles of the granular material. Other investigations used tracer "zones" rather than tracer particles. The tracer zones were composed of the same granular material as the bulk but with a contrasting color [7,8]. As the blade moved through the material, the tracer zones were disturbed and dispersed, allowing mixing mechanisms to be studied.

Movement of particles in a quasi 2D rotating drum was studied by Orpe et al. [9] and Mellman et al. [10]. These studies focused on the effects of drum size, particles size, and rotation speed on layer formation in the 2D bed. Photographs and video recordings were obtained through the transparent side walls. Recently, similar studies were performed by Mandal and Khakhar [11] for mixing of non-spherical particles. The effect of aspect ratio on the dynamic angle of repose and shape of the free surface were studied. Kuo et al. [12] studied the translation and propagation of material bands in the top layers of rotating drums containing binary mixtures at an initially mixed state. Video recordings

were taken from the top of the curved surface and effects of fill level, particle size ratio, and particle elastic properties were reported. Similar rotary drum experiments were performed by Liu et al. [13]. Collected data was then used to compare the speed and accuracy of different image analysis algorithms and regression models in determining mixing times. Development of material bands in a spherical tumbler was investigated by Finger et al. [14]. The spherical vessel was made of transparent material and images were taken from the top. Smaller particles were observed in the central axial band while larger particles accumulated on the sides of the vessel.

Yang [15] and later Tai et al. [16] reported the effect of bed height and vibration intensity on segregation in a vibrated bed containing steel and glass beads. Imaging systems were employed by Litster et al. [17] to observe flow regimes in a vertically bladed mixer. “Bumping” and “roping” regimes were identified. In the bumping regime, granular media moved up and down with the passage of the blades. In the roping regime, granular media was thrown to the periphery of the vessel and centrifugal forces dominated. Hu et al. [18] studied flow in a rotating conical vessel, with and without stirrers. The vessel’s shape and opacity did not allow for wall imaging. In both cases images were taken from the top free surface. Knight et al. [19] studied the convection of granular materials under vibrations. Dyed tracer particles were added at known material depths contained in a cylindrical vessel subjected to vibrations. The time taken by the tracer particles to rise to the surface indicated the dependence of the initial particle depth on convective flow. A unique optical technique was employed by Liu et al. [20] where an additional thermal infrared camera was employed along with a regular RGB camera. Red and white plastic spheres were used in the mixing experiments where white particles were held at room temperature whereas red spheres were heated to 40°C and 80°C. Mixing was then performed in an oblate spheroid shaped drum and the process was imaged using both the RGB and thermal infrared cameras. Similar to other optical methods, the thermal camera method was also found to suffer from the limitation of being able to provide data at the free surface or container walls only.

Qualitative imaging techniques are low cost and well suited for preliminary investigation of granular mixing, surface phenomenon, flow regime transition, and pattern formation. For example,

mixing of glass beads and red oak chips in a transparent continuous screw mixer was investigated by Kingston and Heindel [21]. Four different cameras were employed at four different projections to capture the entire periphery of the mixing process simultaneously. The qualitative effects of rotation speed and screw pitch on mixing effectiveness and particle velocity were reported. However, without post processing such as image analysis (particle image or particle tracking velocimetry), much of the data regarding the flow mechanism like velocity profiles (of individual particles as well as tumbling motion and behavior with respect to collisions with each other) cannot be quantified. Inherently, data acquisition is only possible near walls and free surfaces. In cases where moisture or electrostatic forces cause particles to stick to the walls, even this becomes challenging.

While direct visualizations offer a wealth of knowledge, much of it is qualitative or focused mostly on the mixing effectiveness or segregation patterns in layers of material and not on individual particles. In order to gather more information, especially particle trajectories and velocity profiles, it is imperative to employ advanced procedures. Particle Image Velocimetry (PIV) employs the acquisition of consecutive images taken at a known time difference which are then correlated to obtain displacement and velocity vectors of particles in the investigated region [22]. The PIV system requires an illumination system that is focused on the field of interest, a camera that obtains images of the investigation window and a synchronizer mechanism that controls the timing of the image acquisition. Images thus obtained at known time differences are then processed to improve image quality, and reduce noise. Particle velocity is then calculated from particle displacement between the two consecutive images which are taken after a known time delay. In some cases, the investigation window is divided into smaller cells and the light intensity in a particular cell at an instant is cross correlated with the light intensity in neighboring cells at another instant. Using this method, the average movement of groups of particles can be obtained in the form of average particle displacement vectors [22].

Velocity profiles in a quasi 2D rotating tumbler were obtained at the transparent side walls by Jain et al. [23]. A number of studies [22,24–27] were performed on vertical axis bladed mixers. High speed CCD cameras were employed to acquire PIV data at the top free surface as well as the

transparent walls. Effects of blade speed, moisture content, and particle roughness were reported. Data obtained were also used to validate DEM simulations. In studies dealing with wet granular material, image acquisition at the walls was found to be increasingly difficult due to granular agglomeration. Effect of internal and wall frictional properties on blade torque were measured by Darelius et al. [28]. Disc impeller granulators were studied by Khaliltehrani et al. [29] and Reynolds et al. [30] who obtained images at the top free surface only. Radl et al. [31] investigated the flow around a single blade translating in a quasi 2D rectangular cell and reported the effect of blade angle and bed height on the velocity profiles. Granular Couette flow was studied by Hsiau and Jang [32] in a shear cell with a fixed ceiling and a rotating floor. Particle tracking was employed to investigate the effect of wall velocity and solids fraction on the velocity profiles. Scale up effects of impeller speed and fill levels in bench and pilot scale bladed mixers were studied by Cavinato et al. [33]. High speed cameras were installed at the top walls of the bladed mixers and top surface velocities were obtained using PIV.

The major advantage of optical methods is that since the field of optics is highly mature, the equipment is simple and economical when compared to other techniques [28,34]. In most cases, tracer particles only need to be visually different than the bulk material and hence are easily manufactured [6,16]. Various phenomena of interest observed in granular mixing, such as pattern formation and flow regime transition, can be observed at the surface and hence optical methods are well suited for such studies. Being relatively inexpensive, optical methods are also an ideal choice for "pre-experiments", where a phenomenon may be studied qualitatively, and other more accurate techniques can later be employed if necessary. Optical methods typically have high spatial and temporal resolutions that allow these methods to be readily extended to advanced methods such as PIV or particle tracking velocimetry (PTV) [35]. As such, optical methods are found extensively in lab scale systems [36] and easily extendable to pilot scale systems [37]. The greatest limitation of the digital imaging techniques however, is that only wall or surface phenomenon can be investigated. As will be discussed later, certain phenomena at the free surface or near the wall can be substantially different than those in the overall bulk [38]. Additionally, it is not always possible to access the free surface or have transparent walls. This limitation, along with the availability of more accurate "surface

measurement" methods such as spectroscopic techniques, have prevented optical methods from being adopted for large industrial scale applications. Tracer particles could get completely covered by the bulk material at times, causing the location and velocity to not be captured at those instances. The effect of the vessel walls can cause issues such as reflections, while scratches on the wall can affect image clarity and complicate image processing [23]. Due to these limitations, optical methods have not been able to gain further momentum as a technique to study granular mixing in greater detail. This has then necessitated the development of alternative noninvasive methods for the characterization of granular mixing.

Radioactive Particle Tracking

Radioactive particle tracking (RPT) techniques have been well established and find extensive use in various medical fields. The RPT method exploits the disintegration of a radioactive particle to track the location and velocity of the tracer particle in the mixer. Detectors are placed around the mixer vessel being studied and the emission events, detected as a function of the position of the radioactive tracer at a given instant, are recorded. A typical RPT setup is presented in Figure 2 with the data from the detectors being fed to software to reconstruct the path followed by the tracer. It is expected that these particles would follow the trajectory and velocity of the granular media inside the investigated vessel and provide useful information. There are two general types of RPT methods: those where individual particles are activated to emit radiation in all directions (radioactive decaying particles), and those where particles are manufactured to emit photons which occur in two directions 180° apart.

Radioactive Decaying Particles

Tracer particles can be tagged using radioactive isotopes (i.e., Iodine 128, Fluorine 18, Molybdenum 99, Cobalt 66, Scandium 46, etc.). As the isotopes decay with a known decay rate and energy, they emit radiation in all directions that can be detected with a specially designed detector array. Three different reconstruction methods have been discussed by Chaouki et al. [39] by which RPT data can be used to triangulate the location of tracer particles in different types of multiphase equipment. The first method requires the calibration of the individual RPT detectors such that the relationship between the tracer particle's counts recorded on the detector with its distance from the

detector is established as a polynomial function. The second method uses a Monte Carlo mapping procedure that relates the response of each detector with a spatial location within the mixer vessel. This method takes into account both the effects of vessel geometry as well as the characteristics of the attenuating material [40], whereas a third method utilizes neural network models to reconstruct the particle position.

Radioactive particle tracking was employed in a Nauta mixer by van den Bergh [41] to study residence times and flow velocities in different regions of the mixer using Iodine 128 as the tracer particle. A Scandium 46 tracer was employed by Doucet et al. [42] to study the velocity profile in a V-blender and results were used to validate simulation models. Alizadeh et al. [43] studied the effect of polydispersity on mixing and segregation of glass beads in a rotating drum mixer. Different sized glass beads were used to perform monodisperse and polydisperse mixing experiments. In both cases, a few of the glass beads (that contained Na^{23} as soda lime) were prepared in a nuclear reactor to contain the Na^{24} isotope that allowed the activated, yet identical, glass beads to be used as tracers. Little difference was found between the velocity profiles of the mono and polydisperse cases. The smallest particles were observed to congregate in the core of the bed whereas the largest particles accumulated at the periphery.

Roy et al. [44] discuss the different strengths and limitations of RPT when applied to a gas-solid riser. On a single plane, at least 3 detectors are needed which should be distributed evenly around the vessel. Tracer particles must also be chosen such that the source strength is sufficient to be detected by the detectors placed outside the vessel (i.e., the signal doesn't get attenuated too much within the vessel). An exceedingly high source strength however, could saturate the detector leaving the equipment unable to respond to the signals.

One of the greatest challenges of the radioactive particle methods is that, typically, only a single particle can be tracked at a given time. However, a few specialized extensions of the RPT tracking algorithm have been developed such that multiple particles can now be tracked. This requires the radioactivity of the different tracer particles be considerably higher or lower than the other particles. Rasouli et al. [45] used a multiple radioactive particle tracking (MRPT) technique to study the

movement of cylindrical particles in a rotating drum. The orientation of the particles was obtained by affixing, and then tracking, different radioactive particles at either end of the cylindrical particles.

Positron Emission Particle Tracking

The most popular radioactive particle tracking technique utilized in granular flows is called Positron Emission Particle Tracking (PEPT). The radioactive particles chosen for PEPT undergo a beta decay event and emit positrons. These positrons interact with the electrons in the medium around them and are annihilated, producing a pair of photons that are emitted back-to-back (180° apart) with a known energy (e.g., 511 keV for ^{18}F isotope) as shown in Figure 3. If both photons can be detected, the position of the trajectory on which the particle that gave rise to these photons lies can be determined [46]. If a line is drawn from the detection points of the coincident photons, the tracer particle would lie somewhere on this line which is termed as the Line of Response (LoR) [47]. Multiple LoRs then should intersect at the location of the tracer particle. In most practical cases, some of the detected events are "corrupt" such that they either suffered scattering before being observed at the detector, or a pair of coincident detections are not a result of the same annihilation event. In such cases the corrupt readings are rejected and the location of the tracer particle is detected based on iterative triangulation. As the particle moves through the system, its location is continuously tracked and a Lagrangian path and velocity of the tracer particle can be recreated [48].

Similar to MRPT, Yang et al. [49] utilized three particles moving at speeds as high as 240 mm/s and could follow the particles' trajectories. The required algorithm worked similar to a single particle tracking algorithm, with the major difference being that when the strongest tracer particle was detected, photons originating from all other tracer particles were rejected as anomalies, however once the first particle has been detected, the trajectories associated with that particle were removed from the iterations and the next particle was located, with the photons originating from the next weaker particles being rejected as anomalous and so on. Tracking multiple particles can have its limitations. For example, if the particles move too close to each other such that the separation distance is lower than the spatial resolution of the camera, individual particles would be difficult to locate. Similarly, if

the particles moved into areas of lower camera sensitivity, the camera may not be able to discern between the strong and weak radioactive particle signal [50].

Different types of mixers have been studied using PEPT. These include: horizontal axis bladed mixers, vertical axis bladed mixers [51–55], V-mixers [56], and rotating drums. Studies by Laurent et al. [57,58] traced the velocity profiles of granular media in horizontal bladed vessels and determined the presence of "dead zones". The effect of fill levels and blade angles on the axial and radial velocity profiles were investigated by Laurent and Bridgwater [59]. Data obtained from similar experiments were utilized to validate DEM simulations [60]. Mixing effectiveness in horizontal bladed mixers was studied by Martin et al. [61]. Particle motion in the region close to the blades was studied by Kuo et al. [62] where they defined the area affected by the blade as the active region while the unaffected region was defined as the inactive region. At different operating parameters (i.e., fill level, blade size, blade speed), the size and shape of these regions was investigated. At low rpm, the granular media was found to lift over the blade as a rigid body and then slide down as the blade passed below it. At higher rpm, the granular media was thrown from the blades.

Velocity profiles and mixing mechanisms in vertical axis mixers with disc and blade type impellers were investigated by Knight et al. [51]. Stewart et al. [54,55] performed experiments on vertical axis mixers and investigated the effect of fill level and blade speeds on residence time and velocity profiles. Inactive or dead zones were identified and experimental results were used to validate DEM simulations. A more complex conical frustum shaped, vertical axis high shear granulator was studied by Ng et al. [63] and velocity distributions were reported. Effect of fill level and particle density on the velocity profile were also reported [64].

Rotating drum tumbling mixers operating at low to medium speeds were investigated by Ding et al. [65]. Kuo et al. [56] observed the circulation patterns in V-mixers. Seville et al. [66] used PEPT results to validate DEM models in both rotating drums and V-mixers. The more complex Turbula® mixer was investigated by Marigo et al. [67] and results were compared with simulations. An important improvement to the standard PEPT method was implemented by Yang et al. [49] where

they demonstrated that multiple particles could be tracked by using particles of different radioactivity levels.

A simplified yet faster version of the PEPT method is the 2D PEPI (Particle Emission Projection Imaging) that only takes into consideration gamma ray pairs that are incident perpendicular to the cameras' planes. In this case, it would be obvious that the particle being imaged lies in a plane parallel to the plane of the cameras. PEPI is usually employed to study slow moving fluids, particles, and slurries [50]. The PEPI technique was used by Parker et al. [68] to study residence times in a rotating drum.

RPT measurements are a remarkably effective method to track a limited number of tagged particles and have found extensive use in granular mixing characterization. The unique advantage of this technique is that the position and velocity of individual particles can be obtained. Collecting these data over large time periods allows for ensemble averaged spatial and velocity distributions of the granular material to be obtained as well [69,70]. However, as with all "tracer particle" methods, a major challenge with RPT (and PEPT) lies in optimal tracer particle selection. Tracer particles should be able to follow the flow pathlines accurately. Radioactive strength and half-lives need to be selected based on the type of test being run while meeting safety requirements. The most significant disadvantage with RPT is that multiple detectors need to be placed along the entire length of the vessel driving up the cost and complexity of the system. Moreover, the detectors cannot simply be placed around the system randomly and require a careful calibration and optimization procedure before the detectors can be deployed in each new system. Constraints such as mixer geometry and nature of tagged particle often dictate the type, number, and the placement of the detectors employed for a specific experiment [44]. While RPT methods certainly have the capability of becoming an industrial scale technology, RPT equipment is not standardized, requiring each system to be custom designed for the application at hand. Equipment setup and detector deployment is thus a major scale-up challenge, restricting RPT and PEPT to mostly lab and pilot scale applications [71]. Another significant limitation is that imaging multiple tracer particles within acceptable time frames is difficult to achieve. If multiple particles cannot be tracked, particle-particle interactions cannot be captured,

and a smaller number of particles will require very long duration experiments to obtain statistically meaningful data.

X-ray Methods

X-rays are produced when electrons are accelerated and bombarded on a metal target. X-ray methods work on the principle of preferential attenuation of X-rays in the media through which they pass. X-ray measurements are often classified based on the energy they carry. High energy X-rays are more penetrating and are termed as hard X-rays whereas low energy radiation which are readily absorbed are called soft X-rays. Because of their high energy, hard X-rays are used to penetrate in larger and denser objects while soft X-rays are used to enhance contrast [72]. X-ray imaging using common tube sources can be classified into three types: radiography, stereography and computed tomography. Radiography and stereography will be discussed below, computed tomography will be summarized under tomographic techniques.

X-ray Radiography

X-ray radiography produces a two-dimensional projection of a three-dimensional object as shown in Figure 4, which is similar to a dental X-ray. As the X-rays pass through the object of interest, they are attenuated while the transmitted X-rays then produce a snapshot of the attenuation map through the object [72]. These snapshots are obtained either on traditional photographic film or in digital form.

X-ray radiography was used by Mueller et al. [73,74] to calculate the void fraction in a packed bed. The authors were able to quantify the void fraction in both the angular and the radial direction. The pebbles in the bed were manufactured by embedding stainless-steel spheres in the center of Plexiglas spheres. This allowed the location of the Plexiglas spheres to be inferred based on the location of the stainless-steel inserts which preferentially absorbed the X-ray energy.

Rotating drum mixers were studied by Khan et al. [75] using glass spheres and potassium iodide salt particles. The salt particles that were employed absorbed the X-rays more readily and provided the needed contrast for X-ray imaging. Using image analysis, the trajectories of the salt particles were noted. Uchida and Okamoto [76] used tungsten powder as a tracer to study powder flow in a screw

mixer. Powder movement was traced using images obtained from two mutually perpendicular directions. Particle velocity along the transmission direction was found to be one third of the screw speed and this discrepancy was attributed to the high shear force applied on the particles as they moved into the clearance between the walls and the mixer blades. In later works [77], the same authors tested different kinds of screw mixer blades and reported the 2D projected pathlines of the particles as well as the diffusion coefficients.

X-ray radiography is particularly well-suited for applications where specific particles must be identified and located in granular media. Images can be obtained rapidly, and successive images can be used to determine a single velocity component of the artifacts. Image acquisition, storage, and display can be easily automated and current computer power can be readily employed to manipulate and analyze these images [72]. However, the main disadvantage of radiography is that since it only provides a two-dimensional projection, apart from identification of artifacts, little else can be accomplished. Another important challenge is to obtain acceptable image contrast, which is a function of the sample material, X-ray energy, X-ray scattering, and other settings such as the distance between the object and source and detector response time for moving artifacts [78]. Beam hardening is a common error that can arise due to the presence of both high and low energy X-rays, while image clarity and sharpness can also be reduced due to X-ray scatter and penumbra effects [79]. Corrections can be made to account for these errors.

X-ray Stereography

X-ray stereography is an improvement over radiography and requires at least two projections of the object of interest to be taken simultaneously to determine the location of the object as a function of time [80]. Unlike radiography, stereographic methods can obtain two 2D images in quick succession if multiple source-detector pairs are used simultaneously, which can then be processed to produce the 3D map of a particle as it moves through the investigation window. X-ray stereography can therefore be utilized to obtain the trajectory and velocity profiles of individual particles in the mixing vessel. A two source-detector pair system is shown in Figure 5. Alternatively, in stationary

objects or low speed phenomenon, a single source-detector pair can be used with either the source-detector pair or the object being translated or rotated to get a second projection [72].

A modified bi-planar angioscope X-ray system was used by Govender and co-workers [81–84] to track the movement of tracer particles in a rotating mill filled with plastic spheres. As many as eight steel particles could be detected simultaneously in an automated system. Apart from steel spheres, plastic spheres identical to the rest of the bulk media covered with a layer of silver paint were also used as tracers. Particle trajectories and velocity profiles could be obtained at different rotational velocities of the mixing vessel and particle positions could be identified to within 0.15 mm of the actual location. Experimental data were acquired for a wide range of operating conditions and run times, and were subsequently used to validate simulations.

In order to follow the trajectory of the tracer particles it is important that they can be identified as distinct from the rest of the material in the mixer. Tracer particles can be easily identified only if they attenuate X-rays differently than the rest of the bulk, which usually happens if there is a significant density difference between the tracers and the bulk. This in turn may cause the tracer particles to not follow a representative trajectory or velocity profile. Using particles that are of an unusual geometry can aid in the detection of the tracer particles in some cases, however that increases the complexity of particle detection algorithms while still facing the issue of not following representative paths. Therefore, manufacturing a tracer particle that simultaneously attenuates X-rays differently while maintaining a uniform shape, size, and effective density is a significant challenge in X-ray stereography and X-ray particle tracking velocimetry (XPTV). A number of methods have been devised by various researchers to obtain suitable tracers for XPTV measurements. One method is to maintain the effective density of the tracers by combining materials of different densities. A number of researchers [85–87] developed polyurethane foam with metal inserts for use in reactors such as bubble columns and fluidized beds. The metal inserts made the particles detectable while the foam ensured that the effective density of the tracer particles matched other particles in the bulk. Morgan and Heindel [88,89] prepared tracer particles by soaking wood spheres in X-ray attenuating potassium iodide which were then allowed to dry. Although this method changed the density of the particles

slightly, the experiments were being performed for organic materials and after the potassium iodide treatment the density of the tracers remained within the range as the rest of the bulk. Another method of producing tracer particles from organic material was followed by Franka [90] who painted sections of corn kernels with silver paint to enhance X-ray attenuation.

Kingston et al. [91] employed two sources simultaneously to perform X-ray stereography in a double screw mixer, which improved the accuracy of location detection while considerably reduced the time required to perform the experiment. A binary mixture of red oak chips with glass beads was used while the tracer particle's attenuation was improved by dipping a limited number of red oak chips in potassium iodide and coating them with a layer of silver paint. The Brazil nut effect in a vibrated bed was observed by Morgan and Heindel [88] using X-ray stereography. They reported the presence of convective currents within the vibrated bed that caused larger particles to rise to the surface.

The greatest strength of X-ray stereography is that multiple particles can be detected and tracked in a three dimensional space simultaneously [92]. This is a significant advantage over radioactive particle techniques which require experiments to be run for long durations to obtain statistically meaningful data [60,92]. In X-ray imaging, the particles do not need to be radioactive thereby allowing for greater flexibility and reduced cost in the manufacture of tracer particles. The opacity of neither the mixer vessel nor the granular material itself has any considerable effect on the measurements and data acquisition is reasonably swift to capture most mixing processes occurring in a wide range of mixer geometries. The major disadvantages of the technique lie in the safety requirements. The use of ionizing radiation requires that the experiments be completed in controlled environments by trained operators. Although the number of detectors required is considerably less when compared to radioactive particle techniques, two source/detector pairs are ideally used in stereographic measurements that drive up equipment costs. Similarly, manufacturing suitable tracer particles that follow the bulk material accurately while attenuating X-rays preferentially, can be challenging.

Laser Doppler Velocimetry (LDV)

The Laser Doppler Velocimetry technique is another optical technique that has been used to study single phase liquid flows using tracer particles and had been extended to study solid particle motion. The technique requires two beams of monochromatic collimated laser light to intersect in a region. This causes interference to occur in the intersection region, giving rise to a bright and dark fringe pattern. As particles pass through the interference region, they reflect light with a distinct frequency that can be recorded by a photodetector. Due to the Doppler shift, a measure of the velocity of the particles that moved through the interference region can be determined. The dual-beam laser method measures the velocity in a single direction that is normal to the direction of the fringe plane. Multiple dual-beam laser systems can be used to provide velocity vectors in all three directions [93].

Longo and Lamberti [94] studied the velocity profiles of sand and glass particles mixed in a transparent rotating drum mixer using LDV. The drum was rotated at different velocities in the range of 0-10 rpm. The authors noted three separate regions in the partially filled drum based on the velocity of the particles, including a flowing layer at the surface, an almost static layer at the bottom, and a region of plug flow where the particles follow a circular path.

Darelius et al. [95] used a similar method to measure the velocity profiles of granular media in a high shear granulator. Axial and tangential velocity data of the dense powder were recorded with a maximum penetration depth of 4 mm from the vessel walls. The granulator vessel was curved at the bottom that caused refraction of the laser and prevented measurements in this area. The impeller tip was 1 mm below the lowest point from which the measurements were obtained, allowing for the particle velocities in the immediate neighborhood of the impeller to be studied. Laser Doppler techniques require the flow field being studied to be reasonably transparent, therefore regions in the immediate vicinity of the impeller, where the bed density was high could not be properly investigated. Further up in the vessel where the bed density was lower, LDV provided reasonable results.

A major drawback of the LDV methods is the requirement of a transparent window so that laser light can be transmitted to the measurement location. Even then, LDV can only measure velocities near the wall or surface and only a single velocity component is obtained unless more than one set of

lasers is used. Spatially, measurements are greatly restricted and are usually obtained at a single point in the system while the optical setup required is complex and expansive. If all three components of velocity are being investigated, as many as six cameras may be required to capture all the data. This makes the equipment setup complicated and expensive while deployment in most industrial environments becomes increasingly difficult.

SPECTROSCOPIC METHODS

Spectroscopic methods are based on the interaction of electromagnetic radiation with matter. Different spectroscopic techniques have been developed to study mixing and can be classified based on the wavelength range of the electromagnetic (EM) spectrum such as microwave, infrared, near infrared, ultraviolet, and visible. Spectroscopic techniques can also be classified based on the nature of the interaction between the EM radiation and matter. Spectroscopic techniques use the fact that electrons exist in an atomic structure in specific "energy levels". Interaction with electromagnetic radiation may only take place in the form of photons. Photons either emitted or absorbed by the atoms and molecules in a sample depend on the difference of these energy levels. Conversely, since atomic structures are unique for different molecules, the wavelength of the emitted or absorbed radiation may be used to characterize and identify the constituents in a sample.

As the EM radiation is incident on the sample, some of the radiation may be absorbed at specific wavelengths. Using specific wavelengths of the incident EM radiation and then observing the spectra obtained after absorption by the sample leads to the quantitative measurement of the constituents in a mixture. Of the spectroscopic techniques that will be discussed below, near-Infrared (NIR) is predominantly an absorption method. Absorption in NIR only occurs at specific frequencies of the radiation corresponding to the molecular vibrations of the constituents. A measure of the radiation in the near infrared range, transmitted through or reflected from the sample, is usually used to calculate the absorption by the sample [96].

Raman spectroscopy is a measure of incident light scattering. Unlike NIR systems, if the wavelength of the incident light does not correspond to any of the energy levels, it can still energize

the molecule to a virtual state depending of the energy of the incident photon. The relaxation back to ground state requires the emission of a photon which is referred to as photon scattering. This scattering can either be elastic (emitted photon has the same energy as the incident photon) or inelastic (emitted photon has a different energy). In inelastic scattering, also called Raman scattering, the shift in the wavelength of the emitted photon can provide chemical and structural information of the constituents of the sample [97].

Fluorescence techniques utilize both the absorption and emission phenomena observed in certain materials. Incident EM radiation is allowed to be absorbed by the sample such that the molecules are energized and move to a higher energy level. However, instead of absorbed radiation, emissions from the molecules as they decay back to their ground state are of interest. Detection of these characteristic photons is used to identify sample constituents.

Near-Infrared Spectroscopy (NIR)

Near-infrared spectroscopy refers to the use of infrared radiation within the wavelength range of 780-2526 nm [98]. When NIR radiation is passed through the sample, it is preferentially absorbed depending on the overtones and combinations of the fundamental frequencies of chemical bonds found within the sample [99]. Most covalent bonds like OH, NH, and CH can be detected in the near-IR range of the spectrum [100].

A typical NIR setup, as presented in Figure 6, consists of a NIR spectrometer which includes a light source, a monochromator, and a detector which detects the light signals that are transmitted or reflected from the sample [99]. Halogen lamps are usually employed as light sources due to their small size, cost, and sturdy design while detector types include silicon, lead sulfide (PbS), and indium gallium arsenide (InGaAs) [101]. Different detectors are selected based on measurement speed and sensitivity to different wavelengths of operation. NIR spectroscopy has been used in both in-line [102–107] as well as off-line [2,108–111] modes for monitoring the mixing process. The advantage of in-line monitoring is that mixing is not stopped for spectroscopic measurements while off-line monitoring requires that the mixer be stopped for samples to be extracted and then analyzed [108]. Most off-line NIR systems are based on diffraction grating or interferometer systems where the

transmittance, reflectance, or transflectance (both reflectance and transmittance) can be measured. While off-line mechanisms offer better accuracy and reliability, in-line systems employ acousto-optic tunable filters for high speed measurements in rugged environments. NIR offers flexibility in the deployment of the probes based on the type of material and process being monitored. Probes can either be fitted into the vessel walls to allow contact measurements [112] or contact-less measurements [113,114] can be performed where the sample passes through the sensors' line of sight without actually touching it. The probes typically fulfill the function of both illuminating the sample space as well as gathering the reflected light signal [115]. Data thus collected is then usually fed to a computer for post-processing and statistical analysis.

Once NIR data are acquired, different data processing techniques are applied to interpret the data in mixing homogeneity measurements. The measure of the standard deviation in the wavelength and the time domain gives information on real-time and overall homogeneity in the mixture [102]. Direct observation of the IR-spectra lends to the qualitative measurement of mixture homogeneity while statistical analysis of the variations in the spectra over time provides objective estimation of mixing end points (i.e., points at which maximum or acceptable levels of mixture homogeneity have been achieved) [107]. For homogeneity measurements, the difference in the obtained spectra and the spectra from an "ideally mixed state" can be compared [116,117] whereas the standard deviation of the moving block standard deviation of the collected spectra can also be calculated to quantify mixture homogeneity and identify mixing end points [103,104].

The NIR technique was used by Arratia et al. [109] and Duong et al. [2] on a Bohle bin mixer using samples extracted from the system. Operational parameters such as fill level of the mixer and blender speed were reported to affect the overall mixing homogeneity. El-Hagrasy et al. [110,118] performed similar experiments in V-Blenders. Later studies performed by Bellamy et al. [119,120] tried to quantify the effect of component particle size and concentration on the mixing profile and end points in a bladed mixer. Effects of fill level, loading pattern, and number of axes of rotation on bin-type blenders were noted by Mehrotra and Muzzio [111].

In-line NIR measurements of mixture homogeneity in a Nauta mixer were performed by Berntsson [105,106] who observed unwanted features in interferograms of the moving samples that were not observed in stationary samples. These artifacts were caused by the changes in the observed reflectance as the sample moves in front of the probes. It was shown, however, that the Fourier transform NIR spectrometers were adequate to monitor samples moving at low to moderate speeds (up to 1 m/s). Granular mixing in a Turbula® mixer for different compositions of a 4 component powder was studied by Wu and Khan [121]. A single NIR probe was inserted into the mixer and held at a fixed depth into the bed center. The spectral data obtained was used to monitor the mixing process and assess its end points.

A more powerful NIR method is NIR imaging which extends the traditional NIR technique to provide spatial information as well [122]. The entire sample can be scanned and based on the NIR spectra, the spatial distribution of the sample constituents can be provided. Two basic scanning approaches are used in NIR imaging: in the "staring image" method, multiple scans are obtained of the same sample at different NIR wavelengths. The images thus obtained at different wavelengths are then later combined to give a complete "map" of the species spatial distribution. The second approach, called the "push brooming method", takes in the spatial information and provides the spectral information for each location which, in turn, can then be used to create a complete map of species spatial distribution [99]. An indium antimonide (InSb) focal plane array camera with focusing lens and filters was utilized by El-Hagrasy [123] to perform NIR imaging using six band pass filters based on the absorption bands of lactose and salicylic acid that were mixed in a V-blender. Images were taken from the top of the blender. NIR imaging was found to be a very powerful tool when compared to traditional NIR spectroscopy since, unlike NIR imaging, NIR spectroscopy requires multiple sampling points to give adequate homogeneity measurements. The NIR imaging technique captured a large volume of spatial data and could locate dead zones as well.

NIR spectroscopy detects the chemical composition of the particles and is thus ideal in identifying different species and their distribution in the vessel. A major drawback of the NIR method is the extensive modification needed in the mixing equipment to install the NIR probes. Other disadvantages

include the "localized" nature of the measurements. In in-line measurements, the probe is essentially only measuring the spectra at the probe locality and the results obtained could be different from the overall mixture. Use of more than one probe at different locations in the mixer is encouraged to provide an overall view of mixing in the entire vessel [115]. The placement of the probe(s) must be done very carefully and prior knowledge of the mixing mechanism is essential [124]. Spectra obtained from the NIR technique are also susceptible to variations in physical characteristics such as particle size [119], packing density [102], and humidity [110]; as a result statistical corrections are needed to cater to these errors.

Laser Induced Fluorescence

The basic principal of Laser Induced Fluorescence (LIF) states that on excitation by a specific wavelength of light, certain materials emit radiation at a different yet known wavelength. Measurement of this emitted radiation can be used to determine the presence of the emitting species. Lai et al. [125] utilized LIF to study blend homogeneity in a 20 ML glass micro blender. Experiments were performed using an argon laser on a drug called Triamterene that gives off peaks at 526 nm and 561 nm when excited by 488 nm radiation. This property was used to evaluate the composition distribution of Triamterene in the bulk mixture. It was observed that increasing the concentration of Triamterene in the investigated samples also increased the LIF signals showing a dependence of the LIF signals on the actual amount of the constituent. LIF signals were observed even at minute concentrations of the drug as well (on the order of 0.1%); this could be particularly useful in the pharmaceutical industry where high potency drugs require a relatively low concentration of the active ingredient as compared to the rest of the formulation. The LIF signals were recorded in real time and mixing end points were determined when the LIF response plateaued in the system. A similar study conducted by Lai and Cooney [126] used a portable LIF device that could obtain measurements at different process points in real time to demonstrate the capabilities of the LIF system. The penetration depth of the source required for excitation of the LIF active constituent was found to be limited, suggesting that in cases of "caking" or "dough-forming" materials that could obstruct the investigation window, the capabilities of the LIF would be severely limited. Results were found to be

consistent when different sized mixers were used. It was also demonstrated that homogeneity could be monitored in cases where the active ingredient was not fluorescing by monitoring the spectra from the excipients that would fluoresce.

Mixing in a tote blender was investigated using LIF by Karumanchi et al. [127]. Fluorescent and non-fluorescent particles were mixed and samples were obtained after known intervals from 13 predefined locations in the blender. LIF measurements were then used to identify the mixing end points as well as dead zones in the blender.

Durao et al. [128] studied the concentrations of 5 different constituents in a 31 component multivitamin powder blend in real-time as the blend was fed to a tableting press. Guay et al. [129] monitored blending of pharmaceutical components in a V-blender using a non-contact probe that was affixed to one of the lids of the V-blender. An acceleration sensor was used to trigger the acquisition of the LIF spectra when the mixture completely covered the probe window. While LIF was observed to be well suited for noninvasive in-line monitoring of granular mixing, it was also observed that the presence of moisture, particle size distribution, density variations associated with bulk porosity, pressure on the sample and, more importantly, the presence of pharmaceutical dyes are all parameters that could affect the spectra obtained from the sample. The method therefore requires manipulation or chemometric corrections to provide accurate measurement results.

The major disadvantage of the LIF technique is that only those mixtures with constituents that fluoresce when excited can be monitored. While being sensitive to low concentration constituents may be a potential positive, it also requires that care must be taken to ensure that minor impurities do not affect the final results. LIF also requires modification of the vessel for in-line measurements because it is essentially an optical method that requires a window to access the sample. Even at the window, the LIF depth of penetration is low [126] and the windows must be kept clean to allow for accurate measurements, which could prove to be difficult in the case of cohesive powders or mixtures prone to caking.

Raman Spectroscopy

Another method to study granular mixing is to illuminate it with monochromatic light and observe its scattering. Since different species in a mixture scatter light differently, they produce a distinct scattering spectrum. This spectrum is called the Raman spectrum while the difference between the incident and scattered light is known as the Raman shift. In order to interpret the spectral data, two basic methods are employed: one requires that the Raman spectra obtained from the sample be compared to "control spectra" that are obtained from predetermined "homogenous mixtures", while the other method entails that the features of the Raman shift band be studied independently, and features corresponding to constituents of interest are identified without a priori knowledge of expected spectra [130].

A "mixing bowl" type planetary mixer was used to mix diltiazem hydrochloride and wax pellets by Vergote et al. [131]. The mixer was equipped with an observation window where a Raman probe was attached to obtain in situ measurements. Mixing was not stopped during observations and mixing end points were calculated using the mean square difference (MSD) method in which Raman spectra were obtained and the difference between two consecutive spectra were calculated. A homogeneous mixture was defined as one when the MSD values were reduced to near zero. Repeatability of the experiments was tested and it was observed that the mixing end points were reached within 14-16 minutes and further mixing did not lead to segregation or over mixing. The effect of mixing speed was also studied and it was reported that higher mixer speeds were associated with smaller mixing times.

Mixing end points were also investigated by De Beer et al. [130] and the effect of loading order and blade speed were reported. De Beer et al. [130] used a Soft Independent Modelling of Class Analogy (SIMCA) method to compare the obtained spectra with spectra obtained from a "homogeneous mixture" to determine the mixing end points. They also performed NIR spectroscopy measurements and reported that both the NIR and Raman spectroscopy yielded similar results. Breitenbach et al. [132] performed Raman spectroscopy to study the homogeneity in the composition of a drug that was mixed in a Bohle mixer and later extruded as tablets, melting the constituents

together; 14 different locations were sampled on three different tablets and the active ingredient was found to be homogeneously distributed. Hausman et al. [133] investigated the effectiveness of Raman spectroscopy in cases where one constituent is in very low concentration which is a common occurrence in the pharmaceutical industry. Mixing was performed in a V-mixer with a Raman probe attached for in-line measurements of the spectra. It was shown that the Raman spectroscopy method was adequate in detecting mixture homogeneity when one of the constituents was only 1% of the overall mixture. Being a quick and in-line method for checking mixture homogeneity, Raman spectroscopy could be used with an automated system for continuous monitoring of the mixing process [133].

Similar to other spectroscopic methods, Raman spectroscopy also requires a line of sight to the sample under investigation and may require the installation of transparent windows. Care must be taken in the selection of the probes in cases where the illumination spot size is small as this might give rise to a sample volume too small for calibration [134]. In Raman spectroscopy, signal strength is a function of the particle size and a large variation in the particle size of the detected compounds may give erroneous results [135]. In some cases, based on the wavelength of the illumination, fluorescence may interfere with the Raman spectra, therefore the incident wavelength needs to be selected carefully [132]. Sample preparation requirements for Raman spectroscopy are nearly non-existent and the method can be easily applied in an in-line system to provide real time data on the mixture homogeneity. Data analysis of Raman spectra is usually simpler than other spectroscopic techniques and automation may be implemented simultaneously to control the characteristics of each batch [131]. Additionally, Raman spectroscopy is sensitive to polymorphism and the mixture of a crystalline and amorphous form of the same compound may also be studied [135–137]. Many compounds are much more easily detected, identified and investigated by Raman spectroscopy as compared to other spectroscopic methods, crystalline compounds in particular usually have a better Raman intensity as opposed to amorphous materials [133].

Spectroscopic methods are uniquely suitable for pilot scale systems [120,129] and easily extendable to industrial scale systems [138,139]. As mentioned, spectroscopic systems require that

only the probe is placed in the granular processing system. While modifications are required in the system to install the probes, these modifications would still be fairly inexpensive compared to other systems that require massive and/or complicated equipment. However, as spectroscopic methods take measurements only at discrete locations, industrial scale systems would require a larger number of well-placed probes for adequate monitoring. Fortunately, unlike most methods, power requirements for spectroscopic equipment do not increase drastically as one moves from lab-scale to industrial-scale applications.

Spectroscopic methods usually have high temporal resolution and are able to discern between different mixer species distinctly. However, they are restricted due to their small scale of inspection as compared to the overall scale of the studied sample, especially in industrial scale vessels [140]. One way this limitation is overcome, specifically in the pharmaceutical industry, is by monitoring not just the mixing process but all the processes leading to the final product [99]. In one such case, after the completion of mixing and other processes, high speed NIR spectrometry was successfully deployed to identify the composition of drug capsules on the packaging line at full line speed (12000 tablets per minute) [141].

TOMOGRAPHIC METHODS

Tomographic methods reconstruct the cross-sectional image from either transmission or reflection data collected by energizing the cross-section of the sample from many different directions [142]. Tomographic systems can be categorized as either soft field or hard field measurement systems [143]. In hard field methods such as X-ray systems, changes in the measured property (e.g., X-ray attenuation) in one location of the investigated region do not influence the readings in another location. This means that that the field lines of X-ray attenuation remain straight during measurements and the reconstruction process is relatively simple. However, due to the source strength and detection systems, temporal resolutions are low. Among the hard field techniques, X-ray imaging is usually considered to be relatively safe as they only emit X-rays when in operation and can be controlled

using the input voltage [39,144]. On the other hand, in soft fields such as electrical capacitance tomography, any change in the capacitance in one region of the system will directly affect the measurements taken from other location within the domain. This complicates the reconstruction process often producing multiple solutions that require iterative and optimization methods to obtain the ideal reconstruction [143].

Tomographic techniques are used extensively in laboratory and pilot scale studies, yet have failed to achieve the same popularity in industrial scale applications. The greatest hindrance to their low applicability is the requirement of bulky and cumbersome equipment. Even in the few cases where tomographic methods have been applied to large scale vessels [145], the experimental equipment had to be custom built for the vessel to be examined. Large power requirements, equipment cost when designing for full scale industrial system and the inherent requirement of the mixing species to have distinctly different densities has restricted X-rays from industrial applications. MRI systems in particular are limited by the inner diameter of the main magnetic coil and are mostly suited for specific laboratory scale studies [146]. Only electrical methods such as ECVT have been used in industrial scale granular flow studies, especially in the fluidization community [147]. Even then the scaling up of the probes needs to be done carefully to avoid errors induced due to large sized electrodes [148].

Magnetic Resonance Imaging (MRI)

Magnetic Resonance Imaging (MRI) utilizes magnetic fields to detect the spatial distribution of atomic (usually hydrogen) nuclei in a sample. As presented in Figure 7, on the application of a strong magnetic field, the nuclei in a sample align their spinning axis with the direction of the magnetic field. The protons also precess around the magnetic field at a particular frequency known as the Larmour frequency which depends on the strength of the local magnetic field. Application of radio frequency (RF) pulses interfere with the precession motion and the alignment of the protons with the magnetic field. As the RF signal is removed, the protons relax back to their original state and emit energy that is detected by the MRI equipment. An example of a typical MRI system is presented in Figure 8 with

the different magnets used within the system. The strength and response time of the emitted signal is used to characterize and image the sample [149,150].

Velocity profiles in a cylindrical vibrated bed were studied by Ehrichs et al. [151] using MRI. Poppy seeds were used as bed material because the oil combined within provided the protons that are essential for MRI detection. Similar studies were performed by Knight et al. [19] and velocity profiles of the poppy seeds in a vibrated bed were obtained. Particles were observed to flow upward in the center while they traveled downward in the vicinity of the roughened walls.

Axial and radial segregation of a binary granular mixture in drum mixers were studied by Metcalfe and Shattuck [152] and later by Hill et al. [38,153]. In these studies, only one of the species in the mixer had free protons and was thus MRI-sensitive. This allowed the segregation and concentration distribution of the two species to be easily determined based on differences in image intensity. Images obtained from MRI imaging were compared with the digital images of the surface and it was observed that the segregation in the bulk did not always extend to or correspond to the surface distribution. Yamane et al. [154] used MRI to investigate the shape of the surface layer as well as the velocity of particles in a horizontal rotating cylinder. Results obtained were used to validate computational simulations. Radial and axial distribution in a rotating conical drum was studied by Kawaguchi et al. [155]. Only one species of particles was MRI sensitive and could be imaged, while the position of the other specie could only be inferred by empty spaces in the images. Dry and moist wheat bran were mixed in a rotating drum bioreactor by Hardin et al. [156]. Moist bran was made MRI receptive while the dry bran was not. Moisture was not observed to seep into the dry bran, nor was clumping and agglomeration observed in moist bran over the course of the mixing process.

Void spaces in packed beds filled with glass beads of different diameters were computed by Sederman et al. [157]. Since glass was not susceptible to MRI imaging, the packed beds were filled with water doped with Cu^{2+} ions. This allowed the MRI technique to image only the "void spaces" occupied by the Cu^{2+} doped water. The overall structure of the bed, average porosity, as well as local regions of high porosity were identified. Similarly, Kawaguchi et al. [158] visualized size segregation in vibrated granular beds. Spherical glass beads of different diameters were placed in a cylinder with

larger spheres forming the bottom layer of the bed. The bed was then filled with an ionic solution which was imaged while the glass beads appeared as voids in the images. Images obtained after the application of vibrational motion showed that the larger particles migrated upwards (e.g., the Brazil nut effect).

Using MRI, efficacy of the Turbula® mixer was studied by Sommier et al. [159] and Porion et al. [160]. The effects of rotation speed, number of rotations, and fill level on the overall mixing of sugar beads was investigated. The sugar beads were made MRI-receptive by dipping them in silicon oil. Dead zones were found to exist in an overfilled mixer. Mixing was found to be complete after only a few rotations and that additional rotations were deemed unnecessary.

Mueth et al. [161] studied granular flow in a couette cell. The cell consisted of two concentric cylinders with poppy seeds and mustard seeds confined between them. Flow was established by rotating the inside cylinder. The rotating inner cylinder induced frictional forces on the neighboring layer of seeds and caused them to move. The MRI system was used to measure the velocities of the particles during the couette flow. In a similar study performed by Moucheront et al. [162], mustard seeds were confined in an annular space as well as constrained by the top and bottom walls. The central cylinder was rotated and the effect of roughness of the confining interfaces was noted in the velocity profiles. The effect of the roughness of the rotating surface was considerable whereas the roughness effect of the top and bottom confining surfaces was limited to the overall shear propagation.

Yabuno et al. [163] observed mass flow and funnel flow regimes in a hopper. The mass flow regime was defined when the entire granular bed moved downward through the hopper orifice while funnel flow was when the body of the granular flow at the hopper periphery remained stagnant while the central core emptied out. Kawaguchi et al. [164] were able to image the annulus region but not the fountain or the spout region in a spouted bed. They demonstrated an important drawback of the MRI system which is that flow phenomenon cannot be captured in low concentration flows.

The MRI technique has been used extensively in granular flow applications yet suffers from certain limitations. The most important one is the inability to track materials without free protons. Most scaled down models that are studied in a laboratory environment use granules made of such materials (glass, plastic, graphite, etc.) and thus cannot be imaged using MRI without substantial manipulations. Additionally, most current MRI equipment is expensive, bulky, and difficult to operate and have high power requirements. Temporal resolution for MRIs are usually poor [149] and spatial resolutions are also unexceptional [165] when compared to other tomography techniques.

X-ray Computed Tomography

X-ray computed tomography (CT) is an advanced version of X-ray imaging. Instead of taking 2D projections of a 3D object, CT allows for multiple projections of the object to be acquired in a manner such that individual cross sectional slices as well as the entire 3D structure can be reconstructed [166]. Reconstruction methods vary based on the type of X-ray beam used. A fan bean acquires individual "slices" of the image called sonograms after which either the source-detector pair or the object itself can be moved to get subsequent slices. These slices are then stitched together to produce a single 3D image of the object. Cone beam X-ray tube sources, on the other hand, generate multiple slices in a single scan to produce the reconstructed image [167]. A cone beam X-ray CT system is shown in Figure 9. Generally, X-ray CT has low temporal resolution and each experiment may take several minutes as either the sample or the source-detector pair must be rotated to gather the data required to reconstruct one complete image. CT imaging when used for dynamic experiments, generally produces time averaged images.

Chester et al. [168] used CT imaging to study the mixing in a double cone mixer. The mixer was filled with particles of different size, a small number of which were impregnated with Molybdenum via incipient wetting. This was done to enhance X-ray attenuation allowing these particles to act as tracers. Both axial and radial mixing patterns were studied and the effect of fill level and number of rotations was noted. Celphere particles impregnated with lead acetate were utilized in studies on V-mixers by Yang and Fu [166]. The effect of particle size and fill level on the mixing efficiency were reported. Qualitative results were also reported on the process of die compaction of the particles. Die

compaction is quite common in the pharmaceutical industry where it is used to manufacture tablets. The movement of the particles during the initial phases of die compaction were clearly visible.

Highly detailed information on the initial and final location of the particles in a mixing vessel can be obtained using X-ray CT imaging. Individual cross sections may be observed to identify dead zones, localized segregation, and any other inhomogeneity. However, the actual process of mixing cannot easily be recreated as the phenomenon itself is faster than the process of obtaining a CT scan. Individual particle trajectories and velocity profiles are difficult to obtain using X-ray tomographic methods. This inherent disadvantage of the X-ray CT imaging stems from the requirement of rotating either the object of the source-detector pair which limits the speed of the image acquisition.

Several researchers have sought to remedy this issue as a high frame rate is required to image not just granular mixers but also numerous other important chemical processing equipment such as stirred tanks and fluidized beds. Mudde and co-workers [169,170] utilized multiple source-detector pairs in a fast X-ray CT system to obtain tomographic data in different planes of a fluidized bed simultaneously. Hampel et al. [171], Bieberle et al. [172] and Fischer et al. [173] devised a different fast CT technique using a moving electron beam. The electron beam when focused at different locations on a tungsten target produced X-rays which provided projections of the object from different angles. Compared to the movement of the object or the source-detector pair, the movement of the electron beam is exceptionally swift giving frame rates as high as 10,000 frames per second [172].

To the best of our knowledge, fast CT systems have yet to be employed in the study of granular mixing, however the “dynamic phantom” experiments performed by [171–173] prove that the fast CT methods could be adapted for use in granular mixing applications. In the dynamic phantom experiments, the above-mentioned researchers tracked the motion of three glass beads enclosed in an aluminum cylinder. A centrally located stirrer, rotating at 20 rotations per minutes was used to induce random motion, while a fast CT scan was performed at 1 kHz [173]. It was shown that the system could be reconstructed in high detail and the particle motion could be tracked accurately using this method.

Electrical Capacitance Tomography

Another tomography technique that has found use in chemical reactors and multiphase flow measurements is Electrical Capacitance Tomography (ECT). As the name suggests, the technique is based on measuring the distribution of the electrical capacitance, which is affected by the material volume fraction across the medium under inspection. ECT measurements require two or more electrodes to be placed around the investigated vessel containing non-conducting materials of differing permittivities. Once the electrodes have been set up, they are energized one after the other causing them to accumulate charge which can be detected as current signals. Typical ECT setups require multiple electrodes as sensors, control systems for the sensors, as well as computer hardware for storage and data processing.

Theoretically, ECT has the capability to monitor mixing of multiphase components in two phases as well as three phase flows. ECT imaging has the advantage of being low cost, high speed, simple to deploy and safe [174]. While uniquely suited for three phase flow monitoring, ECT has not been extensively used for granular mixing applications. Homogeneity of powder mixtures flowing through a glass tube was studied by Ehrhardt et al. [175]. Silicon carbide (SiC) and sugar powders were mixed in a static mixer and sensors were placed before and after the mixer to obtain dielectric permittivity for pre- and post- mixing states.

Grudzien et al. [176] studied “silo music” occurring due to granular flow in a silo being emptied in a controlled manner under the effect of gravity. Silo music refers to the dynamic effects and vibrations observed as the granular material flows through it. The silo was surrounded by sets of 12 electrodes and permittivity values were obtained between different electrode pairs. Using ECT, the effects of silo wall roughness, initial density and velocity of the granular material on the vibrations induced in the silo were studied. It was found that the ECT method was appropriate to study the pulsations due to granular flow in a different regions of the silo.

Later, Rimpiläinen et al. [177] studied high shear granulation using the ECT technique. Water was added to dry microcrystalline cellulose powder as the bladed granulator was rotated at different speeds. Two dimensional tomograms showing the permittivity distributions were later compared with

photographic data to check for accuracy of the ECT method. Solid-liquid suspensions flowing through a pipe were studied by Silva et al. [178] using electric impedance tomography and the radial distribution of the suspended particles were reported using conductivity distribution in the pipe cross section.

ECT techniques are uniquely suited for high speed applications. The electrodes can be quickly charged and switched to obtain signals almost instantaneously [39]. Since the signals are inherently electric in nature, automation and control are also easily implemented [179]. Despite these advantages, ECT has not been popular in the characterization of granular mixing due to a number of reasons. As a "soft field" technology, ECT depends on the distribution of a physical property (i.e., permittivity) in the system to transmit and receive signals for measurements. Dependence on physical properties could mean that the changes in the property at one location in the system can affect measurements at another point in the system. Since the material and thus the electric field is not uniformly distributed in the vessel, this leads to a lowered spatial resolution [180]. Due to the same reason, image reconstruction for ECT tends to be a highly complex procedure that can produce multiple possible solutions [143] and often requires iterative optimization methods to obtain accurate results. ECT measurement also require the installation of electrodes in the system which, depending on the type of the mixing vessel, may not always be feasible. The number and size of the electrodes employed for measurements is usually a compromise between accuracy and certainty in the measurements. More accurate measurements require a larger number of electrodes whereas too many or too large electrodes would result in an average capacitance signal that is not significant enough to be measured with certainty [148].

PASSIVE ACOUSTIC EMISSIONS (AE) MONITORING

Acoustic techniques employ the generation, transmission, and reception of energy as sound waves [181]. The velocity, frequency, and amplitude of waves passing through a medium can be measured to provide useful information about the media through which they penetrate. Sound waves are produced as different particles in the mixer collide with each other, the mixer walls, or the mixer blade. In doing so, part of the sound wave is transmitted through the material while a portion is

reflected. These sound waves can be picked up using a sensor or transducer that is usually constructed of piezoelectric materials which converts the acoustic emissions to electrical signals which are in turn amplified and studied [182]. This acoustic technique used in monitoring mixing processes is termed as "passive" acoustic monitoring because the sound signals being gathered emanate from the mixture or system being investigated itself, unlike other techniques where the response of an actively added signal (e.g., laser beams, radioactive particle, etc.) is employed to quantify a system characteristic [183]. In most cases, statistical methods are then used to interpret the acoustic data. Unlike ultrasonic emissions, audible acoustic emissions travel through the air with minimal attenuation [184] and therefore sensors and microphones do not need to be in direct contact with the equipment or material being investigated. Microphones can thus be installed not only at the walls or bottom of the mixer but can also be suspended into the vessel to get the audible acoustic waves as shown in Figure 10.

Acoustic signals are calibrated with different mixing states which are then correlated with the signals emanating from the mixing vessel to predict mixing states and end points [185]. The frequency of beats pattern and the vibrational modes of the acoustic signals have been found to depend on the material properties [186] and size [187] of the particles being mixed [188]. Tilly et al. [189] investigated mixing in a "bowl type" Kenwood mixer. They deduced that the acoustic signals could be used to determine end points of mixing when steady state in the root mean square of the AE was reached. The amplitude of the emissions was also found to be directly proportional to the size of the particles and the mass of the solids in the vessel by Boyd and Varley [188]. In most cases acoustic systems record the sound emitted from the granular mixer and analyze the relationship between the emitted signals and the mixing state.

Early investigations by Leach et al. [186,187,190] were based on the fact that particles striking each other or the vessel walls emit sound signals that could be recorded and analyzed. Analysis of the fundamental and beat frequencies provided information on the size, shape, and material of the different particles in the vessel. This work, thus, proved that the acoustic method was applicable to multicomponent mixtures and that each component could be separately identified.

The use of acoustic techniques is of particular interest in granulation processes. In the granulation process, smaller particles are made to adhere together to form larger multi-particle "granules". Granulation using a high shear mixer was studied by Briens et al. [191] and Daniher et al. [192] using equipment similar to a bladed mixer. Post processing of the AE signals allowed the characterization of different phases, such as nucleation, granule growth, and granule breakage to be identified. A similar study performed by Whitaker et al. [193] showed that the average signal level of the acoustic waves correlated with the particle size in the granulator. Studies by Hansuld et al. [194–196] on bladed granulators demonstrated that the technique holds promise for end point detection of the granulation process [194] and for in-line monitoring of physical characteristics of the mixture components such as particle size [196] and density [195]. As the particles changed in size, density, and shape, the AE signals emanating from their collisions were found to be altered as well.

In later work, Allan et al. [197] noted the effect of various parameters such as the size of the particles, the mass loaded into the mixer, and the velocity of the blade on the intensity of the AE signals. An important drawback noted by the authors was that the AE spectrum appeared to be dominated by the signals obtained from the collision of the particles with the walls of the vessel and not by the collision of the particles with each other. This could lead to the "measurements" being obtained for this interface only and not from the overall bulk of the mixture. Placing the receiving transducer at different locations of the mixture showed that the AE signal traveled through the entirety of the vessel and thus the noted signal was not localized to the transducer location.

Studies performed by Watson et al. [185] noted that the acoustic technique can be used to identify formation of excessively large granules due to over-wetting and thus could also be used to identify faults in the process. A few studies such as that of Crouter et al. [198] and Crouter [183] studied V-mixers using acoustic methods. They reported changes in the AE signal with the mass, moisture content, and surface roughness of the particles in the mixer.

Acoustic methods depend heavily on the physical properties of the particles in the vessel that need to be properly characterized before experiments. Extensive and time consuming post processing is also required. Another limitation is the required "cleanliness" of the signals. While this can be

controlled in a laboratory environment, in an industrial setting where a large number of sound and vibration inducing machinery are running, noise can become a major problem. Location and velocity profiles of individual particles are also difficult to identify. Despite the drawbacks, acoustic techniques are relatively inexpensive and easy to deploy. They do not require the vessel to be transparent nor extensively modified to install the probes. This makes passive acoustic techniques a powerful tool in characterization of granular mixing.

Acoustic methods are heavily investigated at lab and pilot scales, and upgrading to large scale systems appear to be promising. The effect of different scales was noted by Crouter [183] by using acoustic methods on different sized V-blenders. Migrating from lab scale to process scale equipment may actually assist in data collection due to better acoustics and relatively low cost of the additional equipment. Conversely, background noise and the increased distance from the acoustic source to the detector would be factors to consider for before applying acoustic methods to industrial equipment. Another challenge to the large-scale adoption is that acoustic methods can only be applied to systems where appreciable acoustic emissions are produced such that their evolution can be tracked and converted to data that describes the process under scrutiny [188].

ASSESSMENT OF METHODS OF CHOICE

Various noninvasive measurement methods employed to study granular mixing have been summarized. Each method has its own strengths and weaknesses. Image analysis where cameras are employed are limited to wall or free surface effects. This is particularly problematic for the case of bladed mixers, where the physics of the blade-particle interactions are of prime importance, but are not directly observable. Most optical image analysis experiments were performed in laboratory equipment which was made transparent for this reason. If cameras are to be deployed in an industrial equipment, transparent windows would be required. Modification of the vessel would be in most cases the most expensive step in the deployment of this technique. Cameras, except for some very high-speed cameras, are relatively inexpensive and easy to deploy. Post processing usually requires an image detection and reconstruction algorithm that is used to identify the particle and follow its trajectory over time.

The most important impediment to the industrial deployment of RPT methods is the sheer number of sensors required to accurately locate tracer particles in a flow field. Despite this apparent disadvantage, radioactive particles are well suited for laboratory scale experiments, especially if Lagrangian properties such as particle velocities and trajectories are required. RPT methods are also particularly suited to detect dead zones and residence times in various locations of the mixer vessel. While the number of sensors required is high, they are usually affordable and easy to install and operate. RPT techniques are quick and real time monitoring of particle motion is also possible. The use of radioactive particles increases the safety requirements as they need to be handled carefully. Based on the type of detector, tracer particle, reconstruction scheme, and/or algorithm used, calibration methods need to be tailored accordingly.

X-ray equipment is becoming increasingly more accessible and affordable. However, tracer particle selection remains a major challenge as particles with higher densities are required to attenuate the X-rays and provide contrast. This may help in the imaging process but hinders the particles from following the flow accurately. X-rays techniques can be utilized in different “modes” based on the information required from the system. If particle residence time and/or velocities are required, X-ray stereography can be used. This technique would in most ways work like the RPT techniques and give real time data on particle movement.

Selection of tracer particles becomes wider for LDV, however similar to PIV there arises the need to install transparent windows in the mixer vessel. Compared to most PIV methods, LDV accuracy increases but so does the complexity and size of the required equipment. At the same time penetration depth for the measurements is limited and typical LDV measurements capture only a single component of velocity at a time.

Spectroscopic techniques such as NIR, LIF, and Raman spectroscopy are a fast and accurate alternate method to monitor mixture homogeneity. Spectroscopic techniques are usually employed where not just mixture homogeneity but also the measure of the mixture composition is of prime importance, such as in the pharmaceutical industry. As optical methods, these techniques require a clear view of the mixture to be examined. Apart from the need for investigation windows, calibration

requirements are also high and usually representative spectra from the constituent components, as well as predetermined “homogeneous mixtures”, are required to train the algorithms to identify well mixed states. Due to the fast and accurate acquisition of data, spectroscopic techniques are unique such that they can be used both in the off-line as well as the in-line modes. Penetration depths are low however, and in most cases only the sample exactly in front of the investigation window contributes to the spectra. In order to determine not just local but global homogeneity of the sample, the installation of multiple probes is required. A priori knowledge of dead zones and their proper monitoring can help in reducing the number of probes required for accurate results.

MRI techniques can provide quality information on mixture homogeneity and composition and velocity distributions. Particle trajectories can be followed while flow regimes can also be identified. MRI techniques are usually considered safe given proper precautions are taken with the required high magnetic fields. An MRI machine requires highly trained personnel to operate. In experimental settings, the most prohibitive characteristic is the bulky and cumbersome equipment which requires excessive amounts of power. While MRI imaging can provide a wide range of data, data acquisition itself is time consuming and hence is not recommended for high speed phenomena. In industrial settings, deployment of the huge MRI magnet around the mixer vessel is a herculean task, while it is also mandatory that the particles under investigation are MRI receptive which may not be the case in many industrial applications.

X-ray computed tomography, while time consuming, is able to provide detailed data on the spatial distribution and relative homogeneity of the mixture constituents. Compared to MRI, X-ray CT equipment is neither as expensive nor as bulky, however the use of ionizing radiation significantly increases the safety risks requiring experiments to be performed in a controlled environment. Due to the safety requirements and advanced reconstruction algorithms required, X-ray techniques are usually performed by highly trained personnel that could potentially increase the overall cost of the system.

Electrical capacitance tomography is well suited for high speed applications such as fluidized beds and bubble columns. Compared to other tomographic methods, ECT systems are much simpler

to deploy and operate. Automation is also easily implemented. Spatial resolution is less than desirable, and as a soft field technology, reconstruction is complex and usually requires iterative optimization processes.

Passive acoustic methods are simple to deploy and equipment is fairly inexpensive when compared to all other techniques. Tracer particles are not needed and errors arising from such sources are avoided. Unlike optical methods, mixer vessels do not need to be modified extensively to create imaging windows or probe insertion locations. Power requirements for the acoustic detectors and transducers are almost negligible while the transducers themselves are inexpensive. This, however, is not enough to make them viable for use in an industrial environment where other loud equipment other than the mixer may also be running. Vibrations and sounds from neighboring equipment could pollute the acoustic signals one wishes to study. Extensive calibrations are also required to confidently predict the mixing states. Otherwise, statistical techniques are required to interpret the acquired data to signify mixing end points. Particle scale data such as particle velocities and trajectories are not collected with this technique.

A comparison summary of the relative merits and demerits of the various techniques discussed previously is provided in Table 1. Three solid up arrows represent potential merit, one down arrow means potential disadvantage whereas the one up and one down arrow refer to neither merit nor demerit. The methods have been compared based on pre- and post- processing requirements, ease of operation, and other important factors such as power consumption and safety needs.

Image analysis methods require cameras to be placed around the mixer vessel and also necessitate that at least a fraction of the entire mixer vessel is transparent to be able to get meaningful results. Neutral ratings for most spectroscopic techniques are because while the entire mixer vessel does not need to be transparent, investigation windows still need to be installed. Passive acoustics and ECT require extensive statistical and iterative post processing whereas spectroscopic data has to undergo some form of spectral analysis resulting in their respective ratings.

Table 1: Brief comparison of the potential merits and demerits of the different techniques available to study granular mixing.

Parameters	**Image Analysis**	**RPT-PEPT**	**X-ray Velocimetry**	**LDV**	**NIR**	**LIF**	**Raman**	**MRI**	**X-ray CT**	**ECT**	**AE**
Measured characteristic	Color / opacity	Radioactivity	X-ray attenuation	Interference of light	Absorption energy	Fluorescence	Scattered Light	Magnetic field	X-ray attenuation	Capacitance	Sound and vibration
No vessel modification needed	↓	↑↑↑	↑↑↑	↑↓	↑↓	↑↓	↑↓	↑↑↑	↑↑↑	↑↓	↑↑↑
Post processing requirements	↑↓	↑↑↑	↑↑↑	↑↑↑	↑↓	↑↓	↑↓	↑↓	↑↓	↓	↓
Ease of detectors deployment	↑↑↑	↓	↑↑↑	↑↓	↑↓	↑↓	↑↓	↓	↑↓	↑↓	↑↑↑
Equipment cost	↑↑↑	↑↓	↓	↑↓	↑↓	↑↓	↑↓	↓	↓	↑↑↑	↑↑↑
Experimental run time	↑↑↑	↑↓	↑↑↑	↑↑↑	↑↑↑	↑↑↑	↑↑↑	↓	↓	↑↑↑	↑↑↑
Personnel skill level	↑↑↑	↓	↓	↑↓	↑↓	↑↓	↑↓	↓	↓	↑↓	↑↓
Safety requirement	↑↑↑	↑↓	↓	↑↑↑	↑↑↑	↑↑↑	↑↑↑	↑↓	↓	↑↑↑	↑↑↑
Power requirements	↑↑↑	↑↑↑	↑↓	↑↑↑	↑↑↑	↑↑↑	↑↑↑	↓	↑↓	↑↑↑	↑↑↑
Ease of calibration	↑↑↑	↓	↑↑↑	↓	↑↓	↑↓	↑↓	↑↓	↑↑↑	↑↓	↓

The number and placement of the RPT sensors needs to be carefully optimized, while deploying the primary magnet in MRI around the vessel of interest is always a challenge. Unlike MRI, the cost for the actual X-ray system has been on a downwards curve for a while. However, the actual cost of the setup should include shielding and precautionary systems such as sensors and emergency shutdown instruments, which can drive up the cost. Experimental runtimes for most tomographic techniques are obviously high as the time for the rotation/translation of the object or the detector-source pair has to be taken into consideration. ECT is a notable exception because the system itself does not need to be moved and images of the different sections can be obtained by switching between the electrodes electronically.

Among the techniques listed, X-ray methods have been assigned a negative rating for their safety requirements. This is because as ionizing radiation, specific precautions need to be taken such as shielding and reducing the exposure time and magnitude. The operator needs to be careful during the operation and in most cases the equipment cannot be manipulated while a scan is in progress. A neutral rating for RPT stems from the fact that while specific precautions are also required in the handling of radioactive tracers, the amount and strength of the radioactivity is quite manageable as compared to X-ray systems.

A neutral rating in Table 1 does not necessarily mean that the technique is neither good nor bad in a certain way. It could indicate that in some cases the technique is meritorious while in others it isn't. As an example, vessel modification requirements for NIR have been assigned a neutral rating. While NIR methods exist, particularly for contactless measurements, that require minimal modifications to the vessel, there also exist many cases in which investigation windows need to be incorporated into the mixer vessel. A high number of positive ratings also do not suggest that a particular technique is superior to other methods. Table 1 simply offers a comparison of "how" a technique measures what it does rather than "what" it measures. Therefore, the many down arrows in MRI do not indicate that it is inferior to image analysis methods. It simply means that data collection and processing using image analysis methods is much simpler. The fact that the wealth and quality of data obtained from MRI is far better than what could be obtained using simple imaging is not reflected upon in this comparison.

A comparison on the capabilities (the "what" is measured) of each technique is provided in Table 2 where a check mark represents that the technique is able to obtain the mentioned data whereas a saltire represents that it is not able to obtain the desired measurement. The dash represents a neutral rating which could mean that either the technique is able to get partial data, data in some restrictive cases, or that data can be obtained after major modifications in the system.

Table 2: Granular mixing information that can be obtained from various noninvasive techniques.

Parameters	**Image Analysis**	**RPT-PEPT**	**X-ray***	**LDV**	**NIR**	**LIF**	**Raman**	**MRI**	**ECT**	**AE**
Particle Pathline	✘	✓	✓	-	✘	✘	✘	✓	✘	✘
Mixture Homogeneity	-	-	✓	✘	✓	✓	✓	✓	✓	✓
Mixing Endpoints	✘	✘	✘	✘	✓	✓	✓	✘	✘	✓
Velocity Profile	✓	✓	✓	✓	✘	✘	✘	✓	-	✘
Flow Regime	-	-	-	✘	✘	✘	✘	✓	-	-
Particle Orientation	-	✘	✓	✘	✘	✘	✘	✘	✘	✘
Mixture Composition	✘	✘	✘	✘	✓	✓	✓	✘	✘	✘

*includes radiography, stereography, and computed tomography.

Velocimetric techniques are preferable when particle scale data such as particle trajectories and velocity profiles are needed. All such data are usually lost in spectroscopic techniques such as NIR, LIF, and Raman scattering. Conversely, spectroscopic methods can actually verify the chemical composition of the constituents which makes them a powerful tool for the pharmaceutical industry. Measuring mixture homogeneity, which is actually the most important aspect for the characterization of the mixing process can be accomplished by most of the techniques. A neutral rating is given to image analysis and RPT because in image analysis, homogeneity can only be measured at the free surfaces, whereas in RPT homogeneity needs to be inferred following the location and trajectory of a few tracer particles. LDV being a single point measurement technique is able to provide only localized particle scale data and is limited when it comes to the description of the overall homogeneity.

Velocity data are not captured using any of the spectroscopic methods but velocity profiles can be obtained to a certain extent using the remaining techniques. X-ray CT imaging and MRI can capture certain flow regimes using time averaged images. Optical image analysis can obtain flow regimes if the vessel is transparent and the flow regime can be characterized based on surface phenomenon. ECT can capture flow regime data in which the particle phase is sufficiently dense. X-ray methods are perhaps unique in their ability to even capture the orientation of the particles as they move through the system. This information can be considerably useful in the design of bladed mixers and/or hoppers.

CONCLUSION

In summary, the selection of the noninvasive method to study granular mixing depends on the particulars of the experiment. There are many considerations that need to be accounted for before a successful experiment can be designed, the most important being which data are desired. X-ray stereography and RPT for example, are well suited to follow pathlines of individual particles allowing valuable data such as convection currents and velocity distributions to be extracted. Mixing homogeneity, on the other hand, can be obtained with relative ease using NIR or Raman spectroscopy. Other parameters that can influence the selection of the experimental methodology include, but are not limited to: cost of equipment, post processing requirements, required degree of accuracy, time constraints, and the characteristics of the granular media and mixer.

ACKNOWLEDGEMENT

Portions of this work were supported through the Bergles Professorship and Iowa State University.

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List of Tables

List of Figures

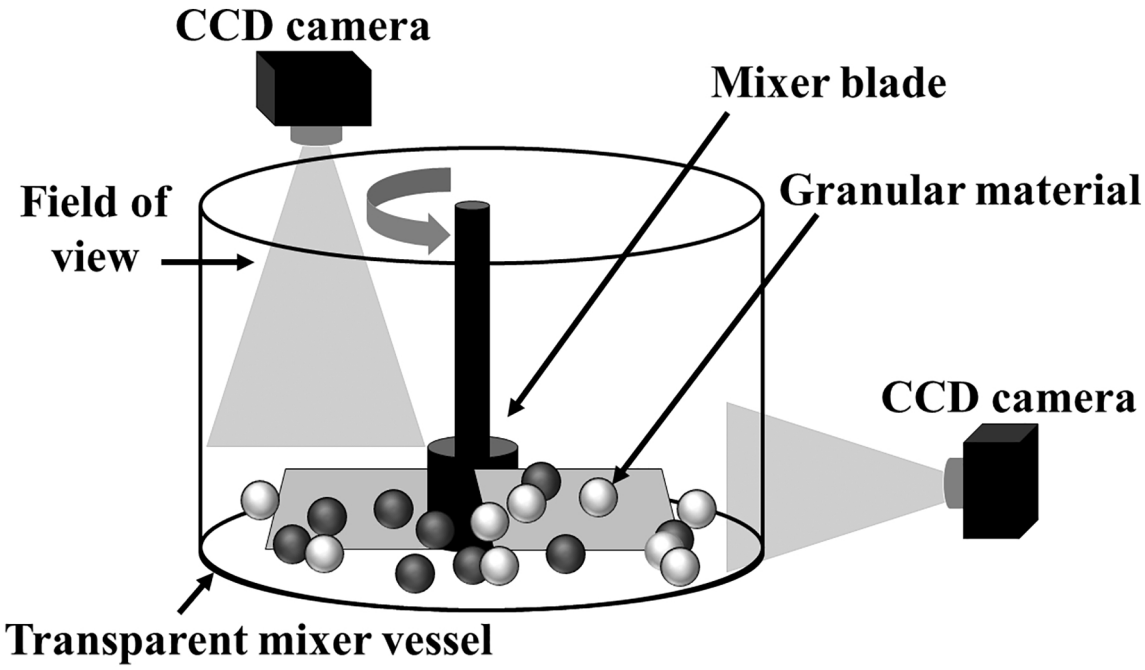

CCD camera
Mixer blade
Field of view
Granular material
CCD camera
Transparent mixer vessel

Figure 1

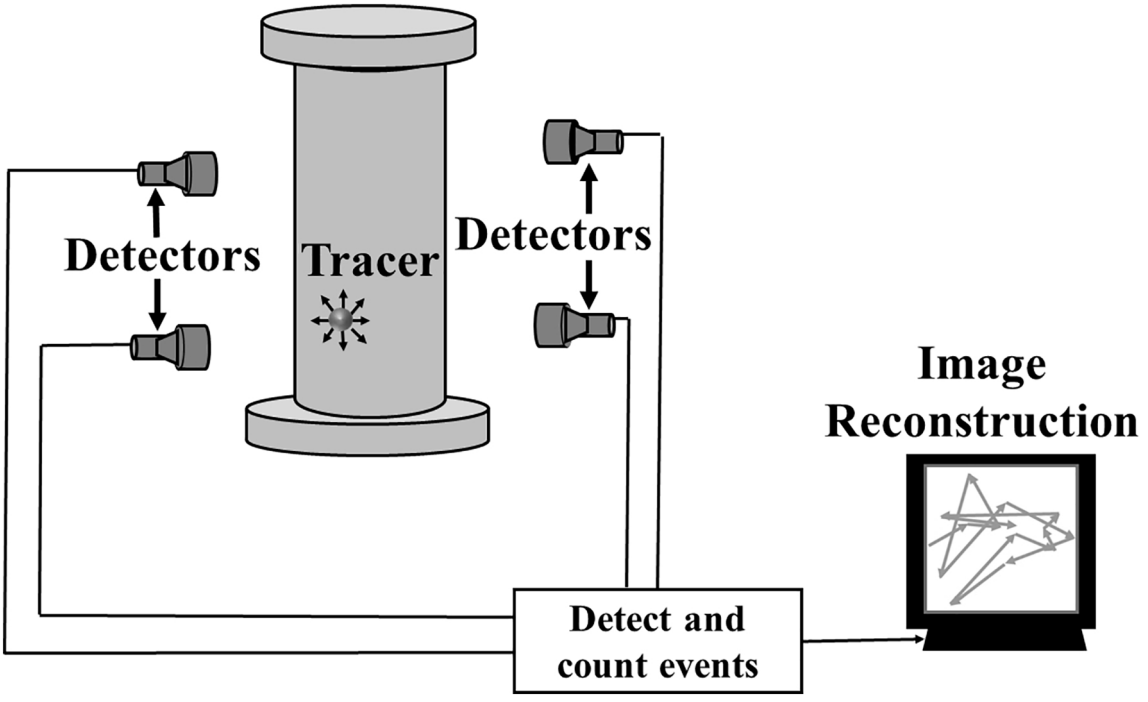

Detectors
Tracer
Detectors
Image
Reconstruction
Detect and
count events

Figure 2

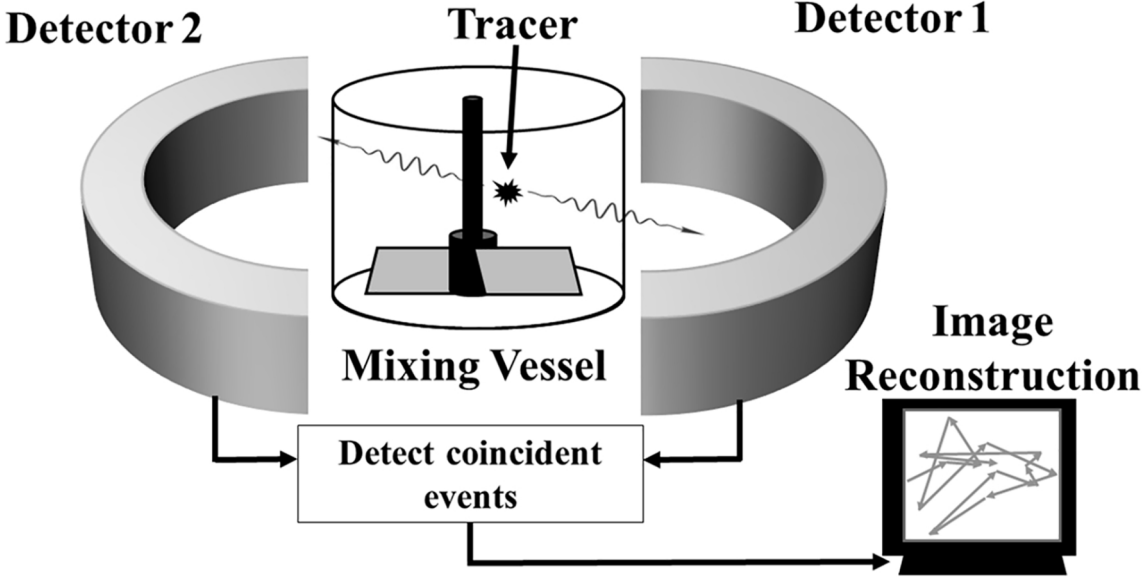

Detector 2
Tracer
Detector 1
Mixing Vessel
Image
Reconstruction
Detect coincident
events

Figure 3

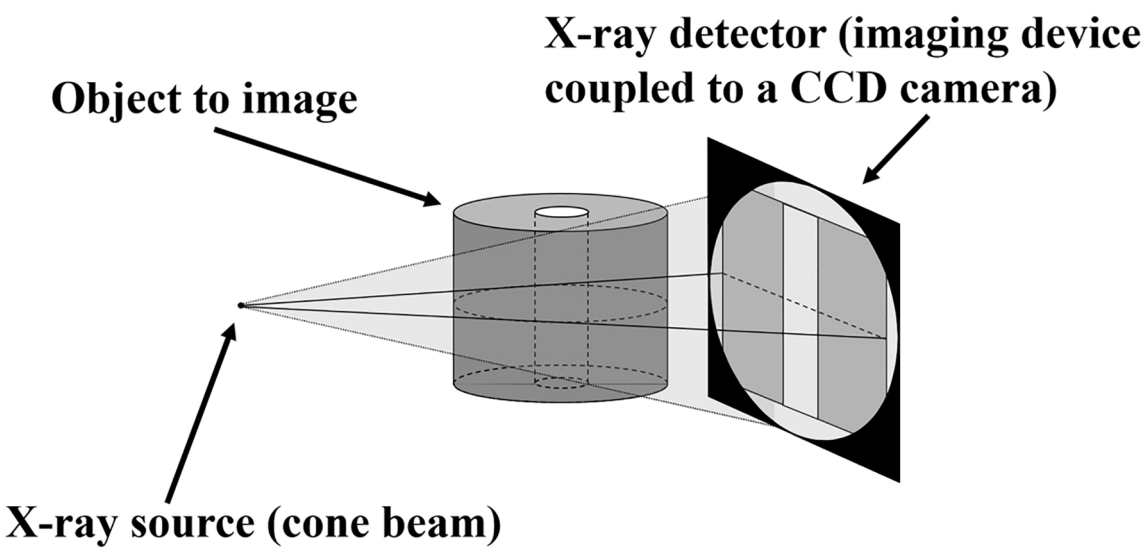

X-ray detector (imaging device coupled to a CCD camera)
Object to image
X-ray source (cone beam)

Figure 4

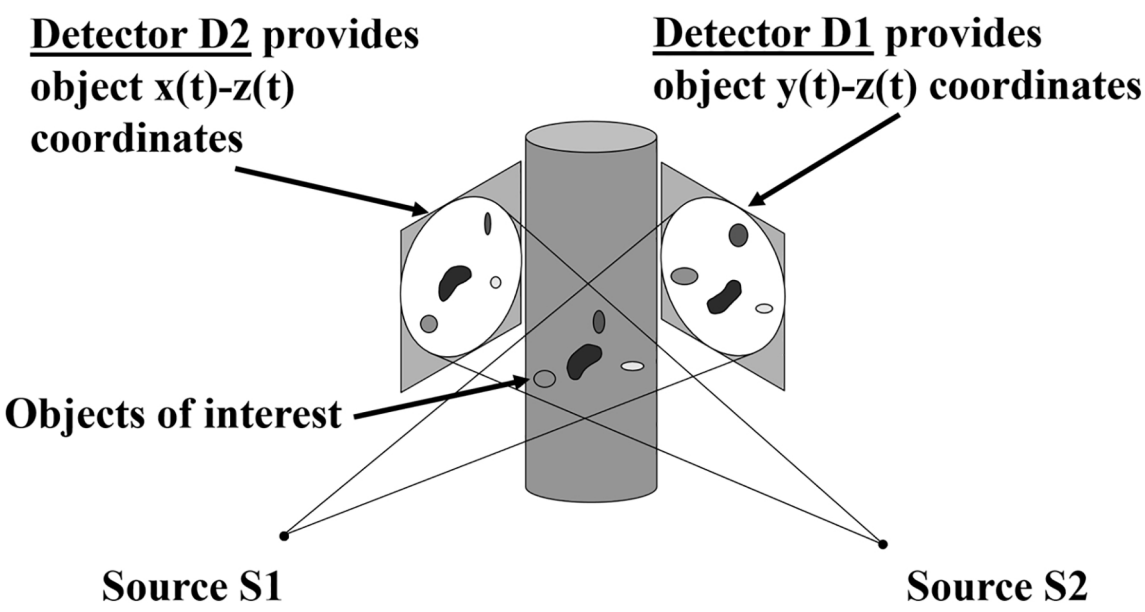

Detector D2 provides object x(t)-z(t) coordinates
Detector D1 provides object y(t)-z(t) coordinates
Objects of interest
Source S1
Source S2

Figure 5

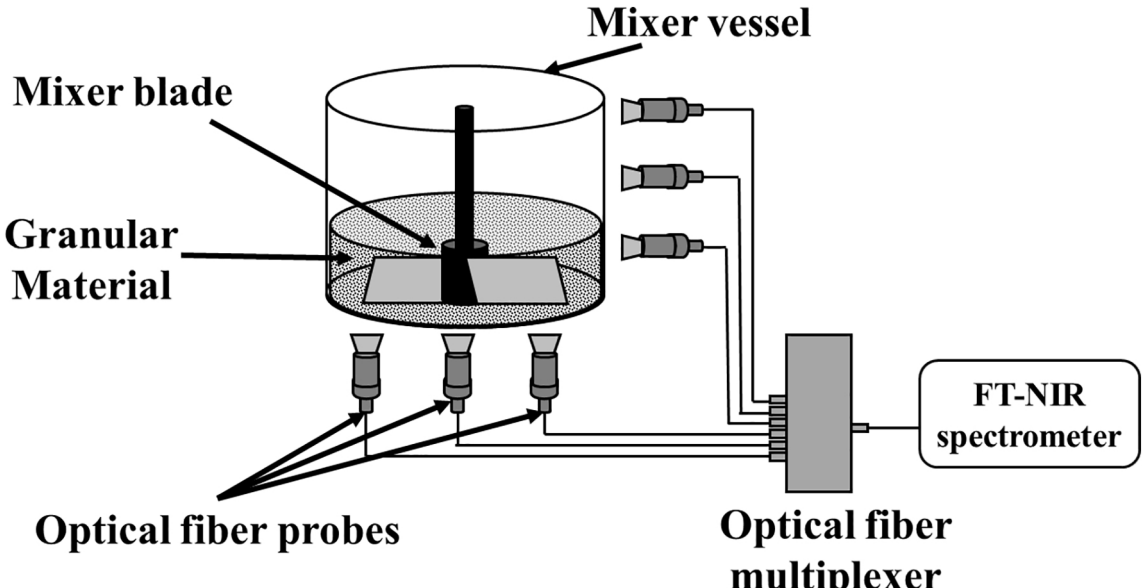
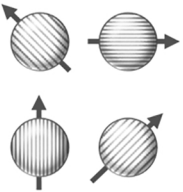
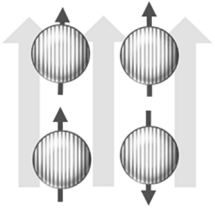

Mixer vessel
Mixer blade
Granular Material
FT-NIR spectrometer
Optical fiber probes
Optical fiber multiplexer

Figure 6

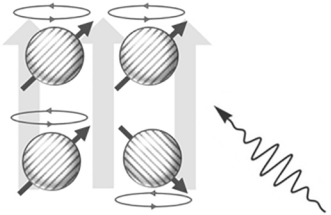
1. Initial Condition – Random Orientation and Spin



2. Spin Alignment with Primary Magnetic Field



3. RF Pulse Applied – Affects Precession and Spin



4. RF Pulse Removed – Protons Release Energy

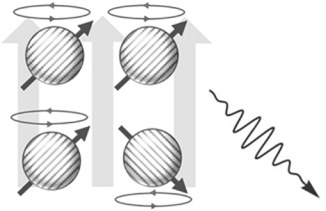


Figure 7

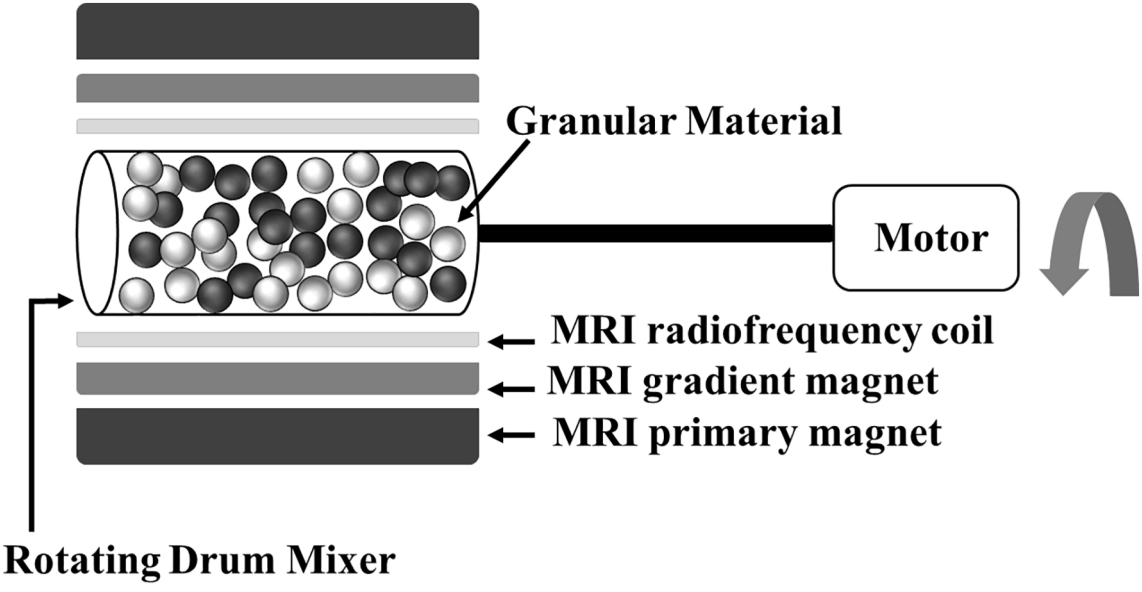

Granular Material
Motor
MRI radiofrequency coil
MRI gradient magnet
MRI primary magnet
Rotating Drum Mixer

Figure 8

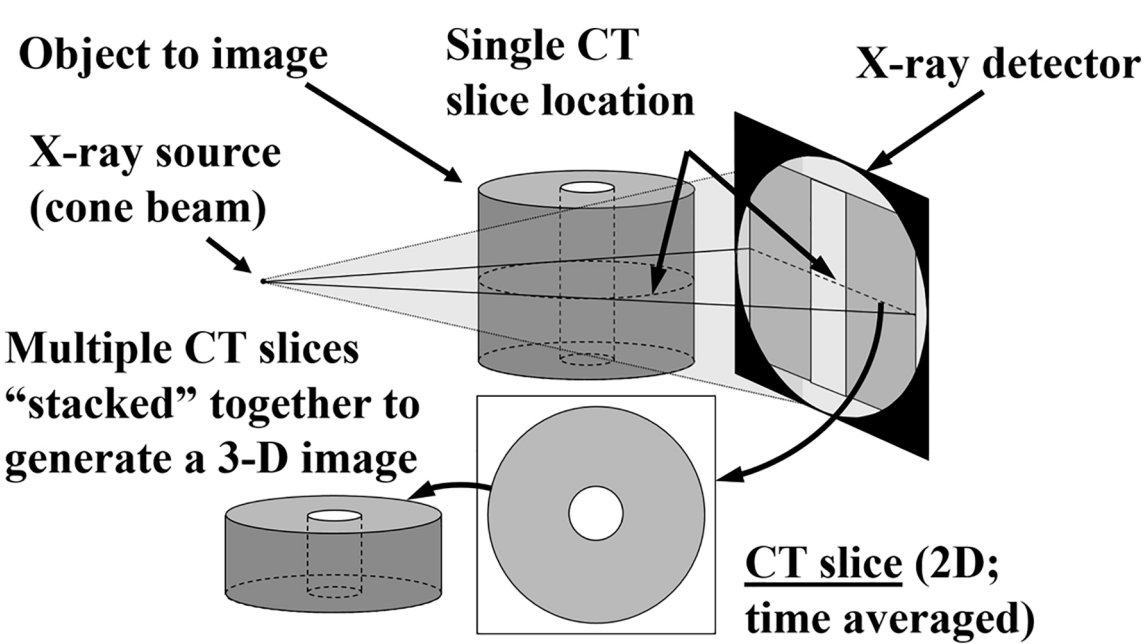

Object to image
Single CT slice location
X-ray detector
X-ray source (cone beam)
Multiple CT slices "stacked" together to generate a 3-D image
CT slice (2D; time averaged)

Figure 9

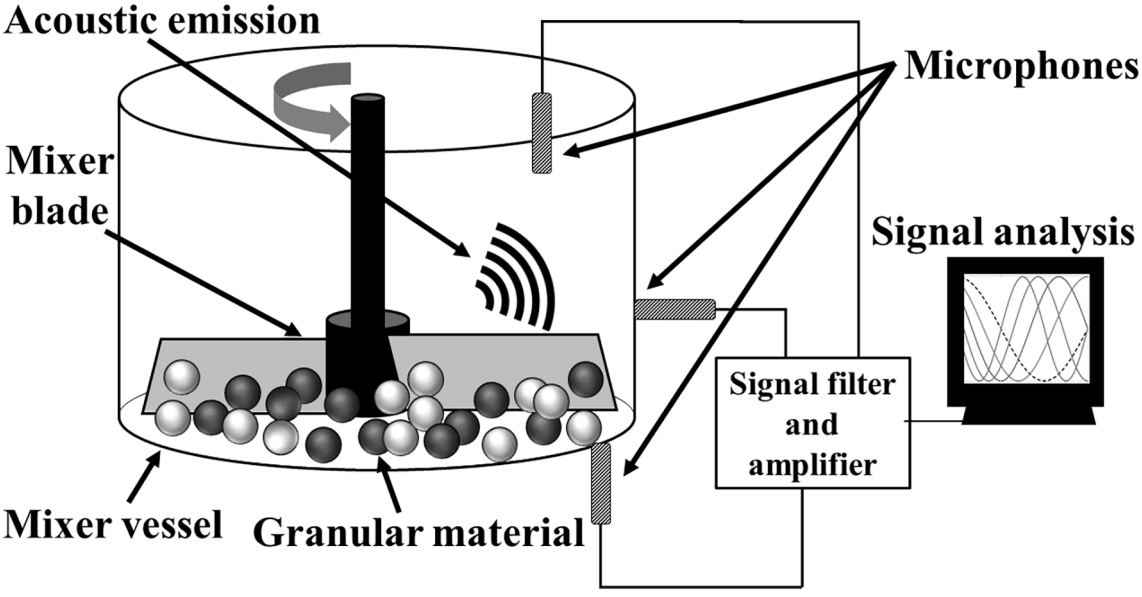

Acoustic emission
Microphones
Mixer blade
Signal analysis
Signal filter and amplifier
Mixer vessel
Granular material

Figure 10