

# Insights into granular mechanics through neutron scattering

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## Abstract

Neutron scattering techniques have provided a window into the structure and mechanics of solid materials for many decades. Recent work by the authors has demonstrated that these techniques can also provide tremendous insight into granular systems. Two prominent examples are presented;

1. The use crystallographic texture measurement techniques to uncover the evolution of fabric within clay soils as a function of deformation, and,
2. The use of neutron diffraction strain measurement techniques to examine the behaviour of force chain networks during the compaction of granular systems.

In both cases, neutrons are able to provide quantitative measurements that are difficult (if not impossible) to obtain any other way. In the first case, this refers to the direct measurement of the average fabric tensor over a representative sample; in the second, neutrons are able to determine the three-dimensional stress state within individual particles and hence allow the calculation of the actual force network existing within an entire assembly of particles.

## 1. Background and Introduction

Neutron scattering techniques have become a cornerstone of materials science and solid state physics for many decades [1]. The combination of penetrating power at wavelengths akin to atomic distances (e.g. 0.5-5Å), and distinctly different attenuation rates as compared to x-rays provide a unique window for the study of crystalline solids.

The vast majority of these techniques rely upon coherent scattering of neutrons as described by Bragg's law;

$$n\lambda = 2d \sin \theta \quad (1)$$

which governs the angle of constructive interference,  $\theta$ , for neutrons of wavelength  $\lambda$ , scattered from a crystal lattice with planes of separation  $d$ , where  $n = 1,2,3 \dots$ . A typical neutron diffraction experiment relies upon the measurement of  $\theta$  for a given wavelength (constant wavelength instruments), or the measurement of  $\lambda$  at a fixed angle (time-of-flight instruments), in each case to determine lattice spacings within a given sample.

A typical experimental setup for a constant wavelength instrument (e.g. KOWARI at the Australian Centre for Neutron Scattering) is shown in Figure 1. An overview of such an experiment is as follows;

From a polychromatic source (typically a fission reactor), a beam of monochromatic neutrons is formed via diffraction within a silicon crystal. This monochromatic beam is then shaped via neutron optics (e.g. slits and/or collimators) to an incident beam that is passed through a sample of interest. Scattered neutrons from this sample are detected, whereby the angle and intensity of the diffracted beam are measured and used to understand the crystal structure.

Two techniques are of particular interest here;

1. ‘Texture measurement’, where the intensity of the diffracted beam as a function of sample orientation is used to map-out the preferred orientations of individual crystal grains within a sample.
2. ‘Strain scanning’, where minute changes in the scattering angle serve to provide a direct measurement of elastic strain (and hence stress) within the material.

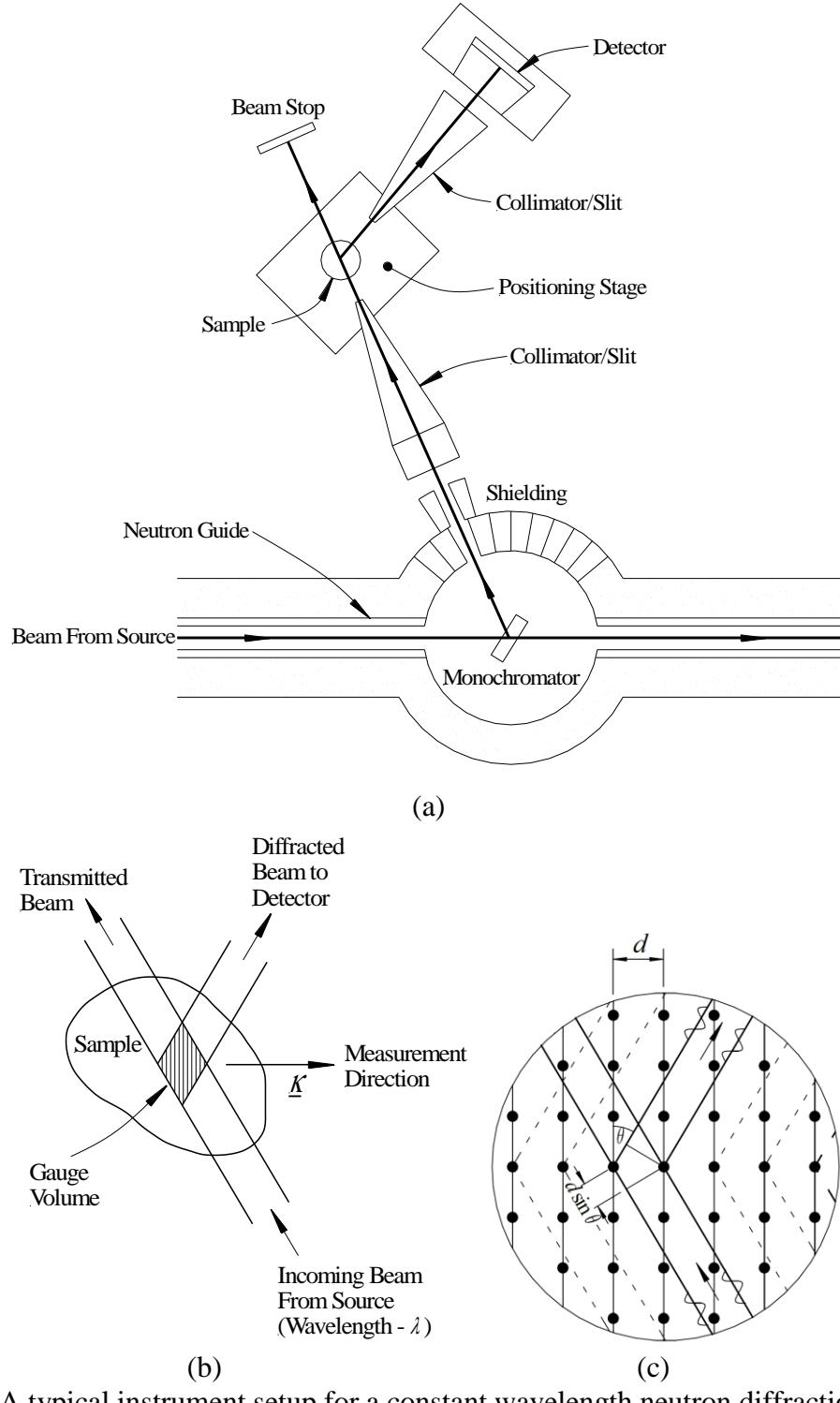


Figure 1: (a) A typical instrument setup for a constant wavelength neutron diffraction experiment.

(b) The angles and intersection of the incident and diffracted beams (as defined by the slits/collimators) define a measurement direction and a gauge volume. (c) Diffraction from crystal lattice planes as governed by constructive interference (adapted from [7])

In recent times, these techniques have begun to be applied to the study of granular systems. Alongside some particularly interesting imaging work (e.g. [2],[3]), this has involved the measurement of the distribution of stress within powders undergoing die compaction [4]-[8], time series investigation of the development of stress within a granular system over a loading cycle [9], measurement of individual particle stresses within 2D and 3D granular assemblies [10], and a fundamental examination of the relationship between granular fabric and plastic strain within clay. In this paper we provide an overview and commentary of this work.

## 2. Texture Measurement and Fabric in Particulate Systems

In a polycrystalline sample, the intensity of the diffracted beam is directly proportional to the number of crystallites that happen to align with the instrument geometry. This simple concept provides a means to map-out the distribution of crystal orientations within a sample. The process is as follows;

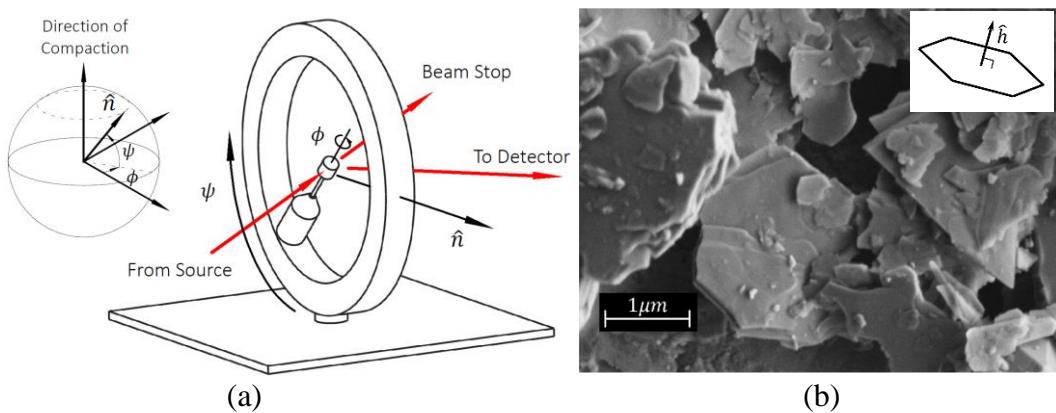


Figure 2: (a) Texture/Fabric measurement on a neutron diffractometer. (b) Plate-like kaolin particles. [12]

With the sample mounted on a positioning system (e.g. a goniometer, or robot arm), the intensity of the diffracted beam is monitored as a function of sample orientation. With reference to Figure 2a, this provides a relative measure of the portion of the sample with crystal planes aligned normal to the direction  $\hat{n} = (\cos \psi \cos \phi, \cos \psi \sin \phi, \sin \psi)$ . Measurement of this intensity over a full grid of angles  $\psi$  and  $\phi$  for a number of different crystal planes allows for the full determination of the orientation density function specifying preferred orientations of crystals within the sample; otherwise known as ‘texture’.

In some particulate systems, crystallographic texture is intimately linked to particle orientation and hence granular ‘fabric’. A good example of this is clay soils. Clay particles (e.g. kaolin) have a plate like nature that is directly related to the layered silicate structure of the mineral (see Figure 2b). The orientation of a particle in space correlates perfectly with the orientation of the crystal structure. From this perspective, a neutron-based texture measurement informs directly on the microstructural fabric of a clay sample. In this case, the full orientation of the crystal structure is unimportant – only the direction of the basal plane is required to define the normal direction of a platelet. This requires measurement of only one diffracted beam from which the Orientation Density Function (strictly a Direction Density Function in this case) can be determined in the form;

$$O(\hat{n}) = \frac{I(\hat{n})}{\langle I \rangle} \quad (2)$$

where  $I(\hat{n})$  is the measured intensity of the diffracted beam as a function of direction. From  $O(\hat{n})$ , standard fabric tensors of arbitrary rank,  $r$ , can be determined of the form [11];

$$F_{i_1 i_1 \dots i_r} = \langle \hat{h}_{i_1} \hat{h}_{i_2} \dots \hat{h}_{i_r} \rangle = \frac{1}{2\pi} \int_0^{\pi/2} \int_0^{2\pi} O(\hat{n}) \hat{n}_{i_1} \hat{n}_{i_2} \dots \hat{n}_{i_r} \cos \psi d\phi d\psi \quad (3)$$

Hence neutron diffraction can provide a quantitative bulk average of fabric in representative samples of material – a task that is difficult (if not impossible) to achieve any other way.

As a simple demonstration of this technique, consider a recent experiment focused on examining the development of fabric during uniaxial compaction of kaolin clay [12]. This experiment consisted of preparing a series of samples compacted to known densities as a function of moisture content. The fabric within each sample was then measured using the distribution of the {0002} lattice planes on the KOWARI diffractometer operated by the Australian Centre for Neutron Scattering (ACNS) at Lucas Heights in Sydney Australia. Results of this experiment are shown in Figure 3 in terms of ‘pole figures’ and the variation in the axial component of deviatoric fabric ( $F_{ij}^* = F_{ij} - \frac{1}{3}\delta_{ij}$ ) as a function of final density and moisture content.

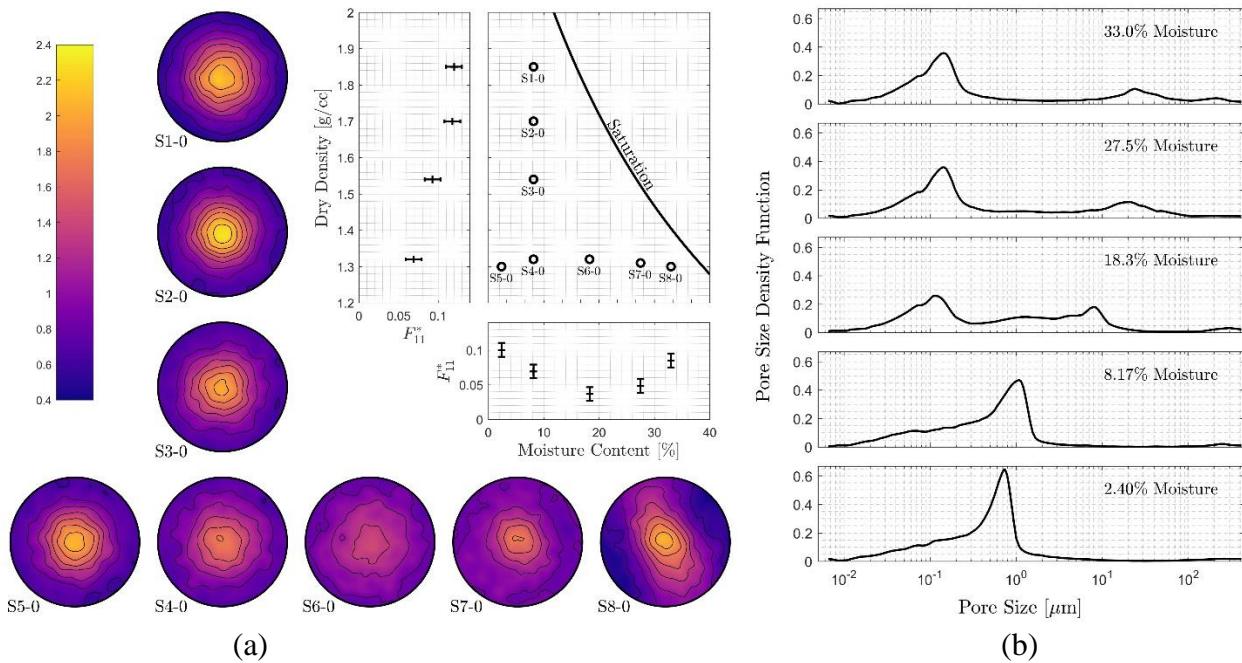


Figure 3: (a) Pole figures showing the three dimensional fabric of compacted kaolin clay samples as a function of density and moisture content. A ‘pole figure’ is a plot of the relative density of platelet orientations on the surface of a sphere as viewed from above a pole. In this case, the direction of the pole is the direction of uniaxial compaction. Deviatoric fabric as a function of density and moisture content is shown in the upper right. (b) The distribution of pore sizes as a function of moisture content via MIP (samples S4-0 to S8-0). [12]

These results show two interesting trends; 1. The preferred alignment of clay particles with the direction of compaction as strain is applied (samples S4-0 to S1-0), and 2. The complex relationship between the development of fabric and moisture content. The latter was examined further through an investigation of the pore structure using Mercury Intrusion Porosimetry (MIP). This indicated a significant change in the pore size distribution over the same range of moisture contents. At low moisture content a mono-modal distribution is observed. As moisture increases, the distribution becomes bi-modal due to aggregation, before becoming mono-modal (dominated by small pores) at high moisture content. Bi-modal distributions are commonly related to random (open) fabrics [13]. In this case it is likely that the local deformation of the aggregates is not purely axial, which would lead to the minimum value in  $F_{11}^*$  observed in Figure 3a.

### 3. Strain Measurement in Granular Systems

Applied stress has the tendency of distorting the crystal lattice within a solid and this has a direct effect on lattice spacing. Modern diffractometers can measure spacing with enough precision to

detect this change and hence provide a means to measure strain. The salient features for such a measurement are as follows;

1. The measured strain is of the form;

$$\epsilon = \frac{d-d_0}{d_0} \quad (3)$$

where,  $d$  is a measured lattice spacing and  $d_0$  is an equivalent reference spacing measured in an un-strained sample. Typical uncertainty is of the order of  $5 \times 10^{-5}$ .

2. The measured strain refers to a single component of normal strain in the direction defined by the bisection of the incident and diffracted beams. By measuring multiple directions (via sample positioning), the full 3D triaxial strain state can be measured.
3. The measured strain refers only to the elastic component. Plasticity is related to the migration of defects and slip systems and has no effect on lattice spacing. This means that Hooke's law can always be applied to diffraction-based strain measurements to calculate stress, regardless of the amount of plastic strain experienced by the system.
4. The measured strain is localised to the region defined by the intersection of the incident and diffracted beams (known as the gauge volume). The size of this region is easily controlled by the neutron optics (slits/collimators). The distribution of strain within a sample can be mapped-out by scanning this region over the sample.
5. The penetrating power of neutrons allows such a measurement to be made deep within a sample (e.g. several centimetres in steel).

Once again, this technique is routine for crystalline solids and has more recently begun to be applied to granular systems. This can provide a window into granular systems from both a bulk point of view or at a fundamental level depending on the relative size of the gauge volume compared to individual particles (see Figure 4).

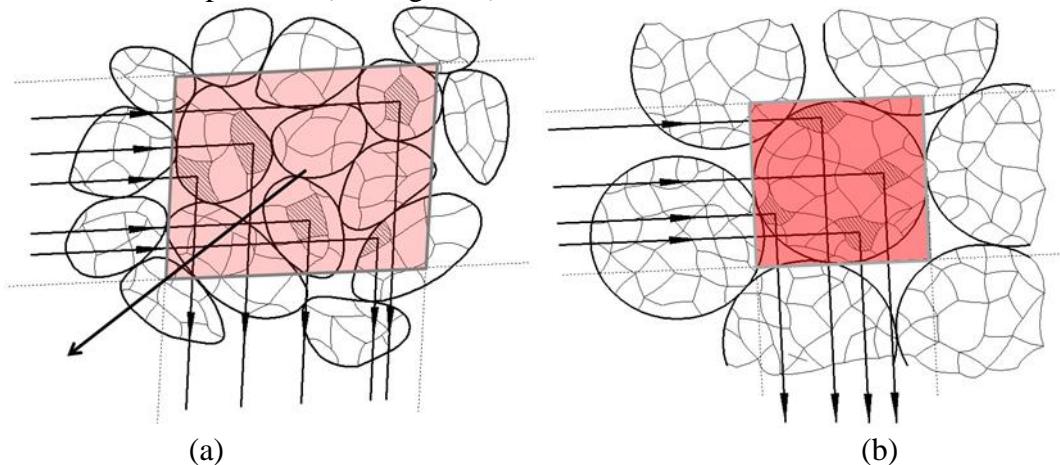


Figure 4: (a) With a gauge volume significantly larger than the particle size, bulk averages of strain (and hence stress) can be made across representative volumes within the sample. (b) Provided accurate positioning information is available, stress within individual particles can be probed using a gauge volume corresponding to the particle size. [10]

In a bulk sense (Figure 4a) the technique measures the average elastic strain over an assembly of particles. Provided this assembly is representative, the corresponding stress (as calculated by Hooke's law) provides a measurement of the bulk stress within the granular material provided an adjustment for the void ratio is made [7].

As an example of this technique, Figure 6 shows the results of an experiment carried out to examine the stress distribution experienced within granular systems compacted within various dies geometries [4]. In each case, a granular sample was loaded to approximately 100kN within a compact die system as shown in Figure 5.

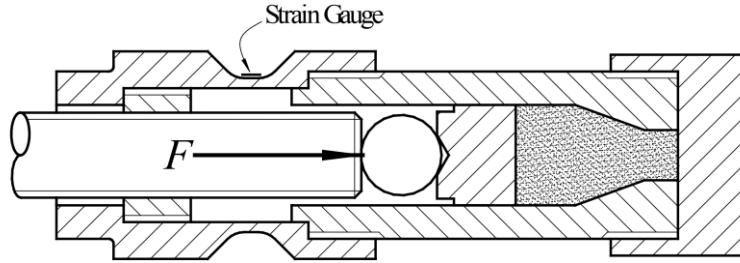


Figure 5: A compact die system for applying axial loads (via the screw thread) to a granular sample.

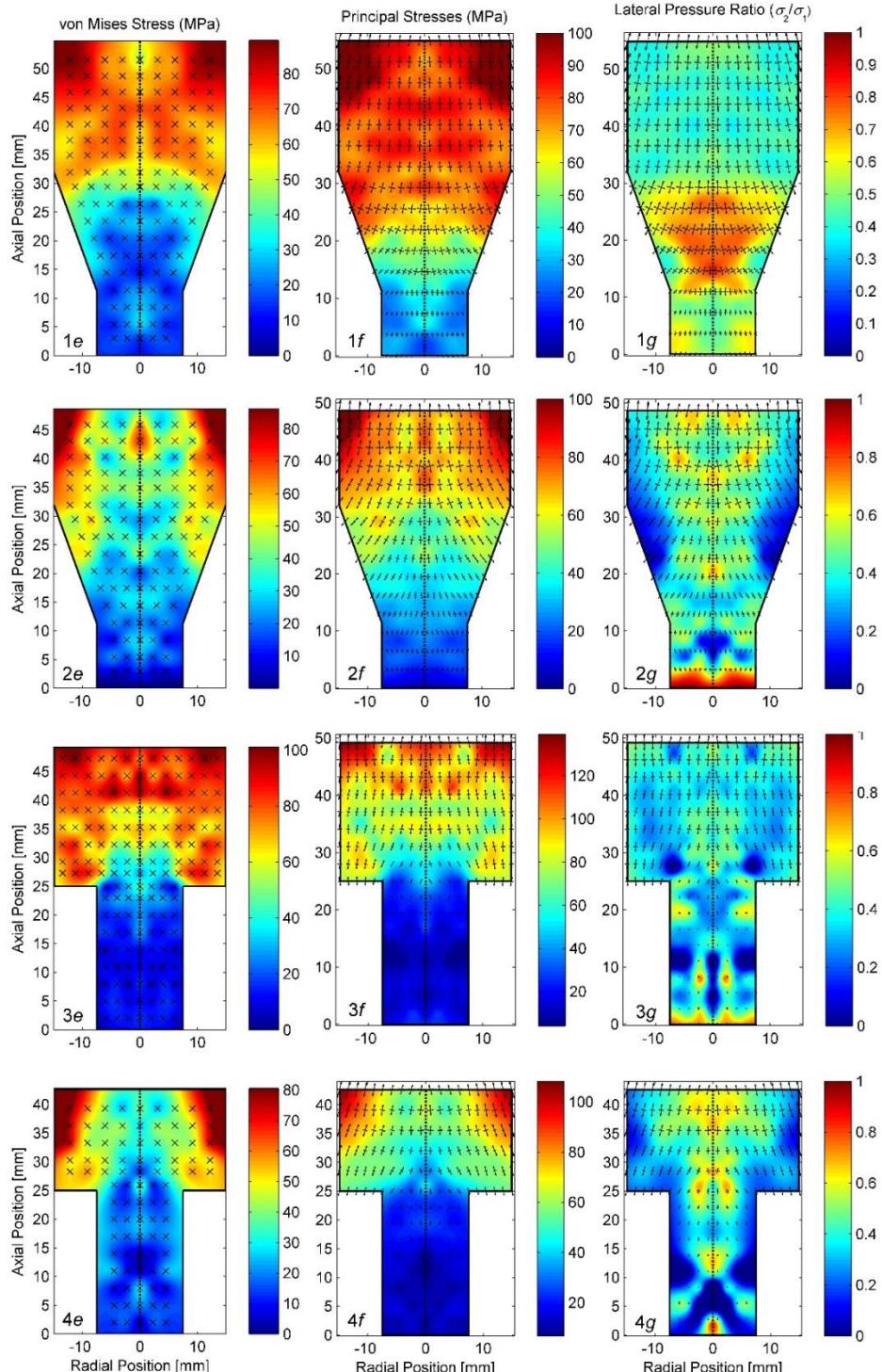


Figure 6: The distribution of axisymmetric stress within granular materials compacted within convergent and stepped dies. Row 1 and 3 – iron powder, row 2 and 4 – quartz sand. Data markers in column 1 show measurement locations, arrows show principal stress in columns 2 and 3. [4]

Two materials; iron powder and quartz sand were investigated within two die geometries; convergent and stepped. After loading, full axisymmetric strain scans were carried out to map the distribution of axial, radial and hoop stress within each system. This was done with a  $3 \times 3 \times 3$ mm gauge volume and the 211 diffraction peak in iron, and a  $4 \times 4 \times 4$ mm gauge volume and the 121 diffraction peak for quartz. Full details of the experiment and its results can be found in [4].

In contrast to this work, neutron based strain measurement can also be used to probe the behaviour of granular systems at the level of individual particles. An interesting example of this can be found in [10]. In this work, a combination of neutron tomography and neutron strain measurement was used to map out the individual triaxial stresses within every particle of an assembly of 549 3.16mm steel ball bearings. This assembly was subject to an applied load of 60kN via the earlier compact loading device (see Figure 7a). In this case, a small device including a Bellville washer and platform was used to penetrate the base of the assembly with a hard post upon loading.

Neutron tomography provided accurate positions for each particle, which were then probed using a gauge volume corresponding to the particle size (i.e. Figure 4b). Measurements were based on the relative change in the 211 lattice spacing in ferrite. In total, 8 components of strain were measured, which allowed the 6 triaxial components of stress to be determined through a least squares process. Some of the results of this experiment are shown in Figure 7b and 7c.

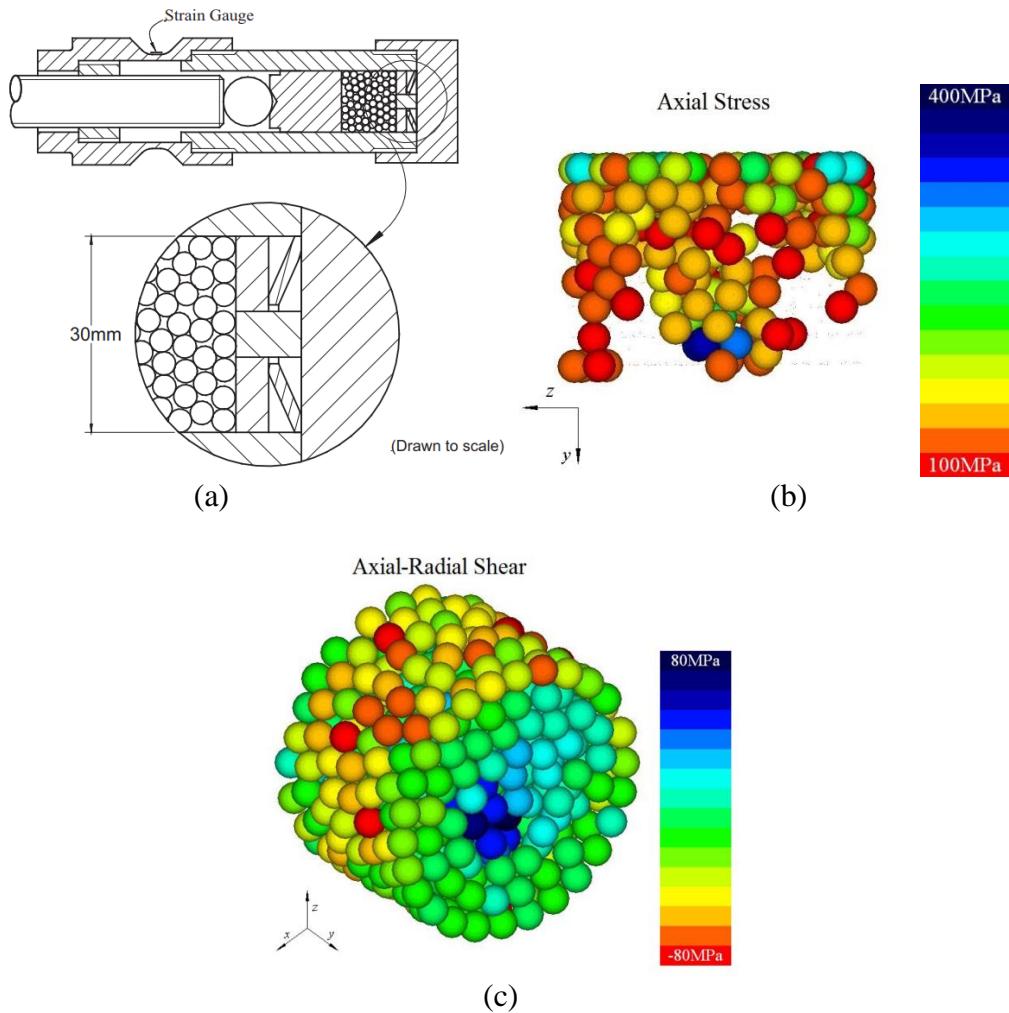


Figure 7: An assembly of 3.16mm steel ball bearings under an applied load within a compact loading device. The base of the device utilised a small Bellville washer to create a hard post that penetrated into the assembly as the load was applied. (b) Axial and (c) shear stress within the assembly as measured using neutron diffraction. (From [10]).

Clear indications of force chains were observed within the assembly, not only emanating from the hard post, but also running up the walls of the die. Detailed statistics of the distribution of particle load throughout the assembly were also available. Full details of this analysis can be found in [10].

#### 4. Conclusions

Established neutron scattering techniques for the study of the mechanics and microstructure of crystalline solids can provide an ideal window into the behaviour of granular systems. These techniques can provide information that is difficult or even impossible to obtain any other way. As shown here, this includes the quantitative and accurate measurement of granular fabric in representative clay samples, the distribution of the full triaxial stress state within compacted powders and the behaviour of granular assemblies at the level of individual particles.

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