The Importance of Rheological Assessment in the Mobilisation, Mixing and Transport of Nuclear Waste Sludges- 11195

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ABSTRACT

The correct usage of rheology data is important in the mobilisation, mixing and transport of nuclear waste sludge from tanks/silos/pond to the treatment plant. Because of their radioactive nature, rheological characterization of the waste sludge is an extremely difficult task. Limited datasets have revealed that these sludges do exhibit thixotropic and shear-thinning, non-Newtonian rheological properties. Because of the scarcity of actual data, recourse has to be made to either making flow curve measurement of simulant sludges having a similar chemical composition to that of the actual waste sludges or assume published rheological data for a given slurry type is applicable for the actual waste sludge. The relevance of using the appropriate shear rate window for pipeline flow or particle support in an unsheared/sheared viscoplastic medium is discussed along with the issues concerning slurry flow curve measurement. A prototype vane rheometer developed by BHR Group that is designed to measure static yield stress at varying and significant depths in sludge beds is also described.

INTRODUCTION

The ultimate disposal of nuclear waste sludge continues to be a highly emotive subject from environmental and political viewpoints. Often called an unsolved problem, it has been considered to be the Achilles' heel of nuclear power [1]. However, attempts to resolve this problem continue to be pursued worldwide. The most viable approach by most countries is the long term storage of radioactive waste after immobilisation (cementation/vitrification) in deep repositories located in stable geological formations.

Most of the waste sludges are currently stored in tanks, ponds or silos located on the same site where the nuclear reactors are situated. The Hanford site in the US state of Washington contains 177 single and double-walled underground storage tanks with 204,000 m^3 (53 million US gallons) of high level radioactive waste. To prevent any further potential for leaks and the subsequent contamination of the Columbia River, there is an urgent need for the contents of these tanks, which were built in the 1940's, to be emptied, immobilised and stored [2]. The requirement for a robust process which can be guaranteed to be safe for several decades has raised significant engineering challenges and finding solutions to some of these challenges has been considered slow. The timescale for emptying these tanks has been put back to 2040 from 2018 [3].

The Sellafield site in Cumbria, UK houses "the most hazardous industrial building in western Europe" (building B30) and the second-most (building B38), which holds a variety of leftovers from the first Magnox plants in ageing ponds [4]. Piles of old nuclear reactor parts and decaying fuel rods are stored under water in the cooling ponds of B30 together with a sludge bed created from the disintegration of contaminated metal into corrosion products over the past few decades. The cooling ponds of B38 consist of radioactive cladding from the reactor fuel rods stored under water along with other undefined material that has been dumped and left to disintegrate. These two concrete edifices are now getting old and their contents must be turned into solid waste which can be contained safely and buried once the UK has decided on the location of a deep underground depository. The clean up bill is estimated to be £50 billion over the next 100 years [4].

The Dounreay site in Scotland used a 65m, 4.6m diameter vertical shaft and a 720m^3 underground silo for the storage of intermediate-level waste (ILW). More than 11,000 disposals of waste were made between 1958 and 1977 when there was an explosion in the head space above the shaft waste after which no further disposals were made. The silo, an engineered concrete-lined bunker, succeeded the shaft in 1971 as a storage facility for the site's ILW with the shaft being used only for items too large for the silo. From 1971 to 1998, the ILW from the site was deposited in the silo through roof ports on more than 16,000 occasions. This waste consisted of failed and redundant equipment, fuel element cans and debris from fuel element breakdown such as cladding, end fittings, wrapper and swarf as well as plastic, glass, paper and filters. The first stage of the clean-up process, namely the minimisation of the flow of groundwater into the unlined shaft using grout to seal the rock fissures around the shaft, was completed in 2008. The next stage, which is scheduled to begin in 2019, is the removal, treatment and storage of the waste from both the shaft and silo. Once brought to the surface, the waste will be segregated and conditioned with cement in 500-litre drums that makes them safe for long-term storage [5].

MOBILISATION, MIXING AND TRANSPORT OF NUCLEAR WASTE SLUDGES

A key component behind the overall process of removing radioactive waste from tanks on the Hanford site, ponds on the Sellafield site or shaft/silo on the Dounreay site is the mobilisation of the consolidated sludge bed. The long storage times of the sludges in these tanks, ponds or silos have caused the sludge to become consolidated, which in turn, led to the formation of high yield stresses. The mobilisation of the sludge bed will require agitation to break up the structure of the consolidated sludge before it can be transported through a pipeline.

Because of lack of moving mechanical parts, pulsed jet mixers (PJMs) are currently being put forward as the best means of agitating the consolidated sludge bed in tanks or ponds [6]. The operating principle of the PJM, shown typically in Fig.1, is two-fold per pulse.

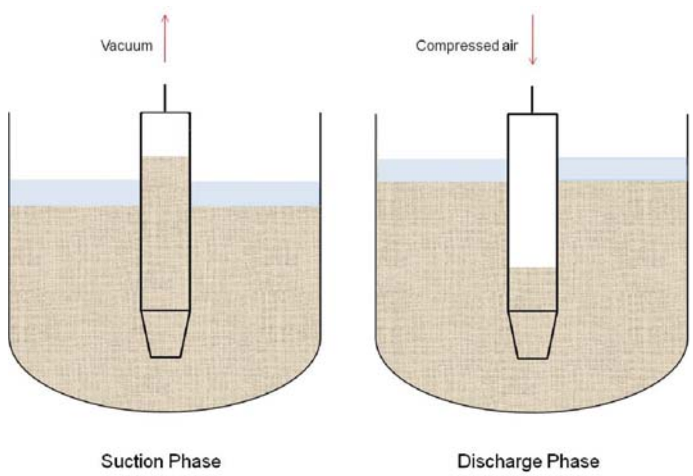


Fig.1. Suction and discharge phases of a typical pneumatically-operated pulse jet mixing system

Vacuum is first applied to break up the sludge in the vicinity of the PJM nozzle and move this sludge into the PJM until a certain sludge height is reached. Compressed air is then supplied to force the sludge inside the PJM through the PJM nozzle. This process is repeated continuously, causing either the structure of the consolidated sludge to break down or the dilution of the consolidated sludge by the incorporation of supernatant water leading to a significant lowering of the yield stress. To avoid the settling-out of the heavy uranium and plutonium particles during the agitation stage and thus any possible criticality, it is vital that careful attention is paid to the rheological requirements necessary for particle support.

Once the sludge is mobilised, it can be pumped through a pipeline to the treatment plant. The sizing and the selection of the pump are both dependent on the pressure drop and volumetric flowrate calculations performed on the sludge flowing through the pipeline. These calculations require the appropriate rheological model parameters for the sludge flowing in the pipeline.

SLUDGE RHEOLOGY

The mobilisation and transfer of the waste sludge from tanks, ponds or silos to the treatment plant requires an understanding of the role of sludge rheology in engineering design. Because of their highly radioactive nature, rheological characterisation of the waste sludge is an extremely difficult task [7]. However, attempts have been made by Sellafield to obtain flow curves of the actual waste sludge in the ponds [8]. These limited datasets have revealed that these sludges do exhibit thixotropic and shear-thinning, non-Newtonian rheological properties. Because of the scarcity of actual data, recourse has to be made with either making flow curve measurements of simulant sludges having a similar chemical composition to that of the actual waste sludges or to assume published rheological data for a given slurry type is applicable for the actual waste sludge.

Despite the poor data acquisition and interpretation, the limited datasets obtained by Sellafield did reveal key rheological information regarding the actual waste sludge. The flow curves obtained were akin to those given in Fig.2 for a thixotropic fluid consisting of a distribution of different types of structure [9, 10]. Fig. 2B shows the existence of separate dynamic and static yield stresses in the same material. This behaviour can be found for bentonite [11], paints [12], waxy crude and fuel oils [13] and many foodstuff products [14]. If the flow curve is explored to very low shear rates with these materials, it is found that the shear stress decreases and then increases with decreasing shear rate, Fig. 2B [15,16].

This behaviour can be explained by assuming there can be more than one type of structure in a thixotropic fluid [11]. Some would be very sensitive and are broken down by the least shear whereas others are more robust and can survive moderate to high shear rates. The robust structure varies with shear rate and determines the equilibrium flow curve (EFC), extrapolation of which then gives the yield stress for the fully broken-down structure At rest, the weak structure will build up over a certain period of time. The combined structure then gives the static yield stress, τ_{ys} which is higher than the dynamic yield stress, τ_{yd}, Fig. 2A. The τ_{yd} is often obtained from a flow curve that does not extend to very low shear rates.

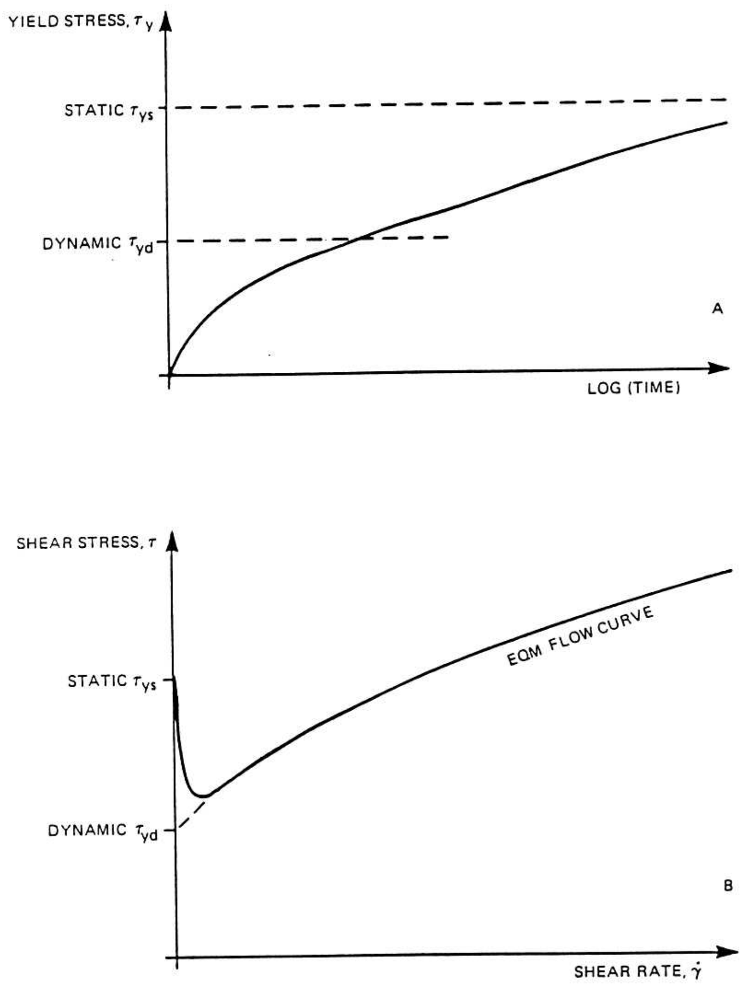


Fig. 2. Static and dynamic yield stresses in multi-structure thixotropic materials

RHEOLOGY AND PIPELINE FLOW

For pipeline flow, the relevant rheological property from Fig. 2 is the equilibrium flow curve with the yield stress being the dynamic yield stress. A model fit should then be made to this flow curve whilst ensuring the shear rate window covered by the viscometer does encapsulate that for pipeline flow. In terms of estimating the shear rate window for pipeline flow, use is made of

$$\dot{\gamma}_w = \frac{1+3n'}{4n'}\frac{8V}{D} \qquad \text{(Eq. 1)}$$

where V is the sludge velocity in the pipeline, D is the inside pipe diameter and n' is defined for each 8V/D as

$$n' = \frac{d\ln\tau_w}{d\ln(8V/D)} \qquad \text{(Eq. 2)}$$

in which τ_w is the wall shear stress in the pipe, defined by

$$\tau_w = \frac{D\Delta P}{4L} \qquad \text{(Eq. 3)}$$

The fit to the flow curve is usually the Herschel Bulkley model given by

$$\tau = \tau_{yHB} + K\dot{\gamma}^n \qquad \text{(Eq. 4)}$$

where τ_{yHB} is the Herschel-Bulkley yield stress, K is the consistency coefficient in units of Pa s^n and n is the power law exponent. This equation describes viscoplastic behaviour when $n < 1$. Because power law ($\tau_{yHB} = 0$; shear-thinning when $n < 1$ or dilatant when $n > 1$), Newtonian ($\tau_{yHB} = 0$ and $n = 1$) and Bingham plastic behaviour ($n = 1$) can be regarded as special cases, the model represents the flow behaviour of a wide range of fluids without being too difficult to handle mathematically.

RHEOLOGY AND PARTICLE SUPPORT WITHIN AN UNSHEARED SLUDGE BED

For particle support within the unsheared sludge bed, the relevant rheological property from Fig. 2 is the static yield stress. This is defined as the maximum shear stress that the unsheared sludge will sustain without developing steady shear flow. By virtue of this yield stress, the sludge bed acting as a viscoplastic medium in the unsheared condition has the capacity to support the weight of embedded uranium or plutonium particles for an indefinite (or sufficiently long) period of time. However, the presence of degraded ILW such as steel drums, paper, glass, etc. may influence the rheological behaviour of the sludge bed in terms of its ability to support high SG particles.

In his comprehensive treatise, Chhabra [17] reviewed the work carried out by several researchers on the gravity settling behaviour of particles in viscoplastic media. For behaviour of spheres under gravity, the parameters governing whether a sphere may settle or remain suspended in the viscoplastic medium are yield stress, τ_{yHB}, particle size, d_p, particle density, ρ_p and suspending liquid density, ρ. At the point of motion/no motion of the sphere, the only two relevant forces are due to the yield stress and the buoyant weight of the sphere. Chhabra [17] concluded that it was convenient to introduce a dimensionless group that is a measure of the relative magnitudes of these two forces. Neglecting all arbitrary constants, the simplest form of a yield-gravity parameter is given as

$$Y_G = \frac{\tau_{yHB}}{g\,d_p\left(\rho_p - \rho\right)} \qquad \text{(Eq. 5)}$$

Published data indicated that Y_G falls into one of two groups, taking values of between 0.04 and 0.08, or a value of 0.212 [17]. The first group, with $Y_G \sim 0.04$ to 0.08, contains the numerical solutions of the equation of motion, experimental results on the observation of motion/no motion in free fall, the measurement of the residual force after cessation of the fluid motion and the extrapolation of force-velocity data to zero velocity. The second group, with $Y_G = 0.212$ contains the postulate of equating the buoyant weight of a sphere to the vertical component of the force due to the yield stress acting over the sphere surface and the measurement of static equilibrium of a tethered sphere in an unsheared medium. The five-fold variation in Y_G can be largely attributed to the different values of yield stress obtained from different measurement methods.

The value of the yield stress to be used in Eq. 5 clearly plays a pivotal role in the interpretation of the experimental data on the settling behaviour of spheres in viscoplastic media. The problem of ascribing a value to the yield stress can be largely attributed to the wide range of values obtained from different measurement methods listed in Table 1.

Table 1 Measurement methods for obtaining yield stress

METHOD	DESCRIPTION
1	Extrapolation of the flow curve (actual and model fitted) obtained from controlled stress or controlled rate rotational viscometry beyond the lowest measureable shear rate [10, 18-20]
2	Creep tests using a controlled stress rheometer with conventional measuring geometries [10]
3	Gun (tube) rheometry [10]
4	Using an "imperfection" in viscometers [10]
5	Oscillatory tests using controlled rate or controlled stress instruments with conventional measuring geometries [10, 21]
6	Stress decay after cessation of steady shear flow using a controlled rate or controlled stress rheometer with conventional measuring geometries [18-20]
7	Spring relaxation tests [22]
8	Controlled rate or controlled stress vane rheometry [19-21, 23-27]
9	Cone/rod/needle/sphere penetrometry [28, 29]
10	Squeeze flow rheometry [30-33]
11	Falling ball/needle viscometry [34, 35]
12	Rolling ball viscometry [36]
13	Stability of an immersed body [37, 38]
14	Stability on an inclined plane [29, 39]
15	Drag plate rheometry [40, 41]
16	Slump test [42-45]

Furthermore, insufficient care was taken by several investigators when carrying out yield stress measurements to ensure the results obtained were meaningful in terms of ensuring the shear rate window covered was relevant to that experienced by a sedimenting sphere in a fluid. The surface averaged shear rate for a sphere moving through a fluid in creeping motion is of the order of (V/d) where V is the terminal velocity of a sphere and d is the sphere diameter. Chhabra [17] quotes that the shear rate range for a sedimenting sphere in a fluid is in the range of 2 to 100 s^{-1}.

In most industrial applications, non-spherical particles are more likely to be encountered than spherical particles. There are two problems associated with the prediction of terminal falling velocity that need to be addressed, namely an adequate description of particle size and shape, and that of its orientation during sedimentation. One approach is to fix the particle shape, size and orientation and study the effect of the Reynolds number. This approach has been quite successful for developing drag coefficient versus Reynolds number relationships for simple shapes such as long circular cylinders, circular discs, spheroidal particles, etc. However, the main limitation of this approach is that it offers no clues about the drag of particles of other shapes. The second approach endeavours to develop a general correlation for all possible particle shapes at the expense of accuracy.

Other than the results of Jossic and Magnin [46], no other reliable results are available for the static equilibrium or drag of non-spherical particles in viscoplastic fluids. At low Reynolds numbers, the drag on a cylinder in vertical orientation increases with aspect ratio (L/d) whereas it decreases when it falls with its axis normal to the direction of gravity. Moreover, the critical value of Y_G is strongly dependent on particle shape and orientation. Values reported by Jossic and Magnin [46] are given in Table 2.

Table 2 Critical values of Y_G in viscoplastic fluids

Particle shape	**Critical Y_G**
Cone	0.026
Sphere	0.088
Cube	0.125
Long vertical cylinder	0.150

Chhabra [17] points out that it is not possible to predict the critical value of Y_G for other shapes in viscoplastic fluids.

RHEOLOGY AND PARTICLE SUPPORT WITHIN A SHEARED SLUDGE BED

Settling of particles in a sheared flow field is sometimes referred to as *dynamic settling*. The motion of a particle falling under gravity is not well understood and cannot be reliably predicted. The structure of the fluid has the potential to affect not only the drag experienced by the particle but also its position in the flow field and its orientation.

A particle situated in a simple sheared flow field will experience a force normal to the direction of shear. This lift or Magnus force arises from the pressure difference across the particle and causes it to move in the direction of increasing velocity. Radial migration of particles away from the pipe wall in the upward flow of granular materials transported in water has been observed by several workers including Engelmann [47] and Newitt et al [48]. In both of these studies, the effect was noticed at high flow rates. Radial particle migration in laminar flow fields is considered by Lawler and Lu [49].

As a first approximation for estimating particle support in a sheared sludge bed is to use the dynamic static yield stress in Eq, (5) instead of the static yield stress. This will give the **minimum** condition that will ensure uranium, plutonium or other particles will remain in suspension once the sludge bed is sheared by the PJM.

SLUDGE RHEOLOGICAL MEASUREMENT

The correct usage of the rheology data is important in the engineering design for the transfer of sludges from tanks to the treatment plant. However, rheological characterisation of these sludges using conventional rotational viscometry is a difficult task [50, 51] in addition to the prohibitively high costs associated with the removal and handling of a radioactive sludge sample for viscometric measurement. Figures of £1m per sample have been quoted with turnaround times of many months. Barring the problems associated with the handling of the radioactive nature of these sludges, the issues that make viscometer selection difficult include

- The sludges in the storage tanks, silos and ponds have a range of non-Newtonian behaviour arising from the wide variation in solids concentration across the tank/silo/pond and within the sludge bed. Typical solids concentrations may range from 10 to 60% by weight with sludge bed heights of up to 10m.
- The height of the free space above the sludge bed can be significant. Typically, this can be up to 16m in height.
- The viscometer/rheometer to be used for flow curve measurement must be robust enough to withstand the plant environment.
- Wall slip may occur when the sludge (usually at the higher concentrations) is sheared when making flow curve measurements. This effect gives a resultant wall shear stress at a given wall shear rate lower than expected due to the formation of a thin layer of fluid (caused by the depletion of the dispersed phase at or near the shearing surface) having a viscosity lower than the bulk of the fluid. Conversely, for a given wall shear stress, the measured shear rate is greater than the true shear rate. In coaxial cylinder viscometry, for example, wall slip can be identified when flow curve data obtained with a series of bobs of different radii do not coincide.
- Ensuring the sample under test is representative.

Laboratory rotational viscometers, which rely on rotational motion to achieve simple shear flow, can be operated either in the ***controlled rate*** or ***controlled stress*** mode. In ***controlled-rate*** instruments, there are two methods of applying the rotation and measuring the resultant torque. The first method is to rotate one member and measure the torque exerted on the *other* member by the test sample, whilst the second method involves the rotation of one member and measuring the resultant torque on the *same* member, The rotating member is either at constant speed which can be sequentially stepped or with a steadily-changing speed ramp. The resultant torque is measured by a torsion spring. In ***controlled-stress*** instruments, either a constant torque (which can be sequentially changed) or a torque ramp is applied to the member, and the resultant speed is measured. The more common geometries used in rotational viscometry are shown in Fig. 3. These are used interchangeably with any of the operational modes described. All of these geometries rely on samples being taken from the sludge bed for remote flow curve measurement.

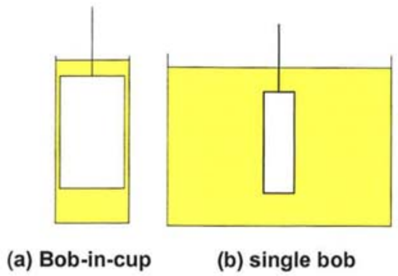
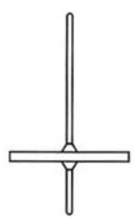
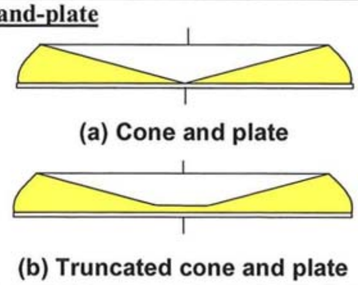
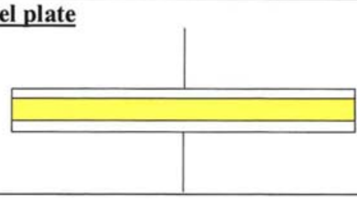
Coaxial cylinder  	The simplest geometry (a) consists of a bob (inner cylinder) located in a cup (outer cylinder) with the test sample contained in the narrow annular gap between the bob and the cup. A variation of the coaxial cylinder viscometer that commonly used is a bob rotating in a large container that approximates to the "infinite sea" situation (that is, the container to bob radius ratio is at least 10).
Rotating disc 	This consists of a disc rotating in a large container (normally a 600 ml plastic beaker with an internal diameter of 97mm) of test material. The torque exerted by the test fluid on the rotating disc is measured as a function of rotational speed.
Cone-and-plate  	Usually, the sample under test is contained between the exterior angle of a cone and a flat plate, (a). The axis of the cone is set normal to the plate with the cone apex touching the plate. Cone angles typically range from 0.25 to 1^{o}. However, for solid/liquid suspensions, a cone with a truncated apex (b) is often used to minimise problems due to particle jamming.
Parallel plate 	The sample under test is held in the gap between two identical circular flat plates. The gap between the two plates can be varied, typically up to 5 mm for plates of about 25 mm radius.

Fig. 3 Types of rotational viscometer

Instead of laboratory rotational viscometers, on-line viscometry could offer a possible alternative for rheological characterisation of the sludge bed. The range of commercial on-line viscometers is diverse but these can be grouped by measurement technique into seven generic types [52]:

- Rotational (coaxial, rotating bob, nutating cylinder and rotating disc) viscometers
- Tube viscometers
- Moving cylinder viscometers

- Drag on float or blade viscometers
- Moving blade viscometers
- Squeeze flow viscometers
- Vibrational viscometers

In terms of the design and its operability with the sludge bed, only the coaxial, nutating and vibrational on-line viscometers appear to be suitable. However, a closer inspection of these viscometers revealed none of these viscometers is actually suitable. For coaxial and nutating viscometers, there was a strong likelihood of particle jamming in the gap between the bob and cup given the wide particle size distribution of the sludge bed. As vibrational viscometers operate at kHz frequencies or higher, this gives rise to a shear wave being rapidly damped down so that the sensor only 'sees' a very thin layer of the sample.

VANE RHEOMETRY

Recent research by BHR Group has focussed on the use of the vane penetrometer that facilitates the direct measurement of yield stress *in-situ* at various depths in the consolidated sludge bed. This approach is based on the assumption that sludges ***do exhibit plastic behaviour*** given that the Bingham plastic viscosities that have been obtained are very small. The vane technique is preferable compared with conventional rotational viscometry because:

- it is an *in-situ* measurement;
- it is a direct measurement of the static yield stress of the unsheared sludge bed, not an extrapolated value of the dynamic yield stress from the flow curve measured using the coaxial cylinder viscometer;
- it can measure yield stress at much higher solids contents of the sludge, well beyond the limit for a coaxial cylinder viscometer.

Previous attempts have been made to obtain *in-situ* yield stress measurements of the sludges in storage tanks using a field vane tester. Because of the significant length of the vane extension rods used, serious bowing and buckling problems were often encountered during the penetration of the vane into the sludge. When this vane was rotated, a significant portion of the rotating shaft was also in frictional contact with the sludge. This led to erroneous yield stress values being calculated on the assumption that the torque contribution of the rotating shaft was thought to be negligible. A prototype was developed at BHR Group to provide a vane rheometer capable of making in-situ measurements of yield stress of radioactive and other sludges as a function of sludge bed depth via computer control. This prototype overcame many of the problems encountered by previous attempts to measure the yield stress of these sludges *in-situ* using a field test tester.

The experimental set-up for this prototype is shown in Fig.4(a). This makes use of the Geonor H60 vane borer supplied by ELE International Ltd. This prototype has been designed to prevent bowing and buckling of the extension rods, which would otherwise result in a nutating action. The vane, shown in Fig. 4(b),with or without its extension rods was rotated at a fixed speed of 6.6 rpm by means of a 24V dc motor coupled with a gearbox.. The whole vane assembly would be lowered into the sludge sample to the required depth by means of a vertical screw jack powered by a 24V dc electric motor with a built-in optical linear displacement encoder. This screw jack has a 1.5m stroke capable of moving at a speed of 50mm/min with a 50kN thrust. Fig. 4(c) depicts the front-end of the control and data acquisition unit that was used for the control of the vertical screw jack and the rotary geared motor for the rotation of the vane, and the data logging of the twist angle as a function of time.

A typical twist angle with time curve is given in Fig. 4(d). As the vane rotates from rest, region A shows an initial transient response, which can be attributed to gear train play of the rotary, geared motor. The linear behaviour at region B originates from the Hookean elastic response of the sample. From the slope of the curve at B, the elastic modulus of the sludge, G, can be evaluated. As the vane continues to rotate, the response becomes increasingly non-Hookean, as shown by region C of the differential angle–time curve. The maximum differential angle given by region D of the differential angle-time curve allows the yield stress of the sludge, τ_y to be evaluated once the geometry of the yield surface and the shear stress distribution on the surface is known. The rapid decline as shown by region E of the differential angle–time curve marks the transition of the sheared sample from a gel to a fluid.

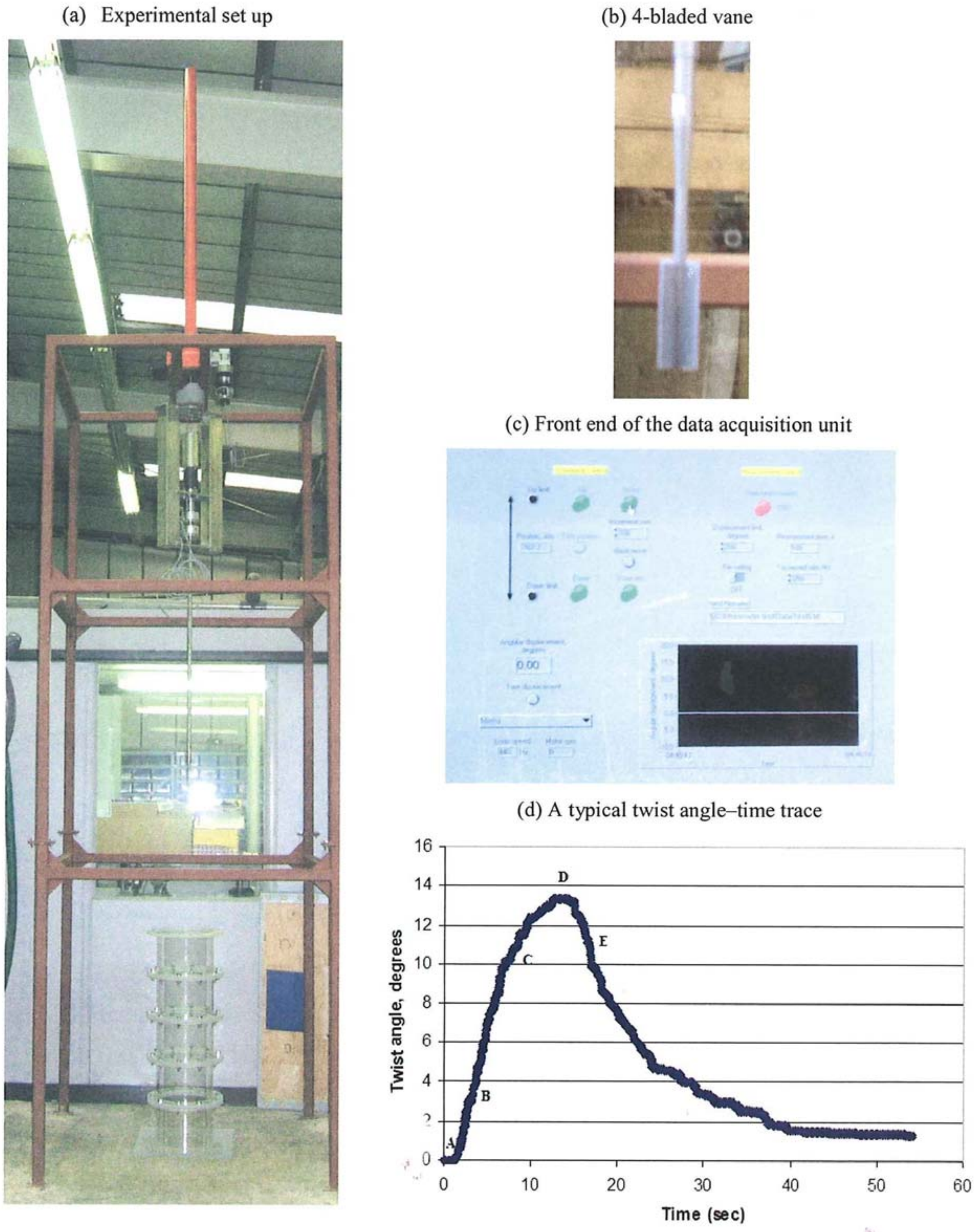


Figure 4 BHR Group Vane Rheometer Prototype

The calculation of the yield stress, τ_y from the maximum differential angle for the *in-situ* vane rheometer is not as straightforward as the laboratory vane tester [23] since the frictional contact between the rotating shaft and the sludge can no longer be ignored. Here, an extension was made to the equation relating the yield stress, τ_y for a fully immersed laboratory vane of diameter D and height H to the maximum twist angle θ_m given by

$$\tau_y = \frac{k\theta_m}{\frac{\pi D^3}{2}\left(\frac{H}{D}+\frac{1}{3}\right)} \qquad \text{(Eq. 6)}$$

where k is the spring constant in Nm/deg.

USE OF SIMULATED SLUDGES OR USE OF PUBLISHED SLURRY RHEOLOGICAL DATA BASED ON SIMILAR SLUDGES

Rheological measurement of simulated sludges is probably the only viable means of obtaining data for engineering design. However, this is not an easy task as information from the actual sludges is extremely limited. An approach that has been adopted is to prepare a sludge which has a similar formulation in terms of its chemical constituents and particle size distribution to that believed for the actual sludge. This is probably satisfactory for sludge flow in pipelines. However, this is not the case for consolidated sludge beds in tanks/silos/ponds as the consolidation process has taken place over several years causing the sludge bed to build significant bed strengths.

Instead of simulated sludges, a desk study of the rheological information published in the literature for various types of slurries as a function of solids concentration, particle size and other physical or chemical properties could be made. Through comparison based on chemical constituent make-up, an indication of the rheological properties of the sludge could be inferred.

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