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Measurement and Modelling of Legacy Sludge Separation and Transport Processes

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ASTRACT

Within the UK nuclear environment, there is an significant accumulation of legacy wastes in the form of particulate-in-liquid 'sludges', contained in an array of ponds, silos and tank storage areas. The largest of these sites is at Sellafield (the UK's largest active nuclear waste facility) where the bulk of hazardous high-priority sludges are designated for separation, transfer and interim storage or final reprocessing within the next 5 – 10 years. Additionally, there are many similar deposits across National Decommissioning Authority (NDA) licence sites, as well as internationally, such as at Savannah River and Hanford in the USA. In response to the major industry challenges associated with removing these wastes, an important Technology Strategy Board (TSB) funded industry-academic collaboration has been formed (comprising Sellafield Ltd, MMI Engineering Ltd and the University of Leeds) that seeks to reduce transfer costs and accelerate process efficiency, by developing two integrated characterisation tools: A novel predictive modelling framework, and an innovative and flexible *in situ* measurement technology. Modelling will allow simulation of separation operations before they take place, giving predictions of efficiencies and allowing performance optimisation. However, because of the complexity of many of the sludge systems, it is imperative that this is linked with characterisation data to validate the model and provide updated performance monitoring. To do this, we are using a novel acoustic backscatter array (ABA) that acts as a high frequency echo sounder, allowing particulate flow concentrations to be measured *in situ* and remotely using the backscattered sound. By combining the power of performance simulation and validation instrumentation, legacy waste transfer will be able to be predicted safely and relatively non-intrusively, leading to essential cost and risk reductions. Presented here will be initial outcomes of the consortium, focused on the design of the acoustic backscatter array and controller hardware. Also detailed will be the design and construction of a 100 litre mixer-settler column, which will be used to calibrate the ABA and for the primary settling trials.

INTRODUCTION

Much of the UK's nuclear legacy waste was generated from fuel reprocessing, effluent treatment and fuel storage during the early nuclear weapons programme of the 1950's and 60's, where government focus to produce plutonium generated secondary waste in amounts that exceeded the industrial capacity to process it. Additionally, the energy crisis of the early 1970's resulted in a ramping up of domestic nuclear power reactors, which again led to waste accumulations from continued bottlenecks in fuel reprocessing operations at that time. This has meant that large volumes of corroded sludges still exist now after 50+ years, these being housed in 'temporary' ponds and silos that are well beyond their original life design. Collectively, they represent the UK's highest nuclear contamination risk, and thus their removal is of national importance. Indeed, it is emphasized that the Sellafield Ltd site represents the single largest UK nuclear decommissioning challenge with an overall projected lifetime cost of £67.5bn as estimated from the NDA's Audit Office report of 2012 [1]. More specifically, over the next 5 to 10 year period (where major waste transfer is scheduled to take place) Intermediate Level Wastes (ILW) and Highly Level Wastes (HLW) held within the site will consume almost 80% of projected costs. Additionally, the economic burden of these wastes hampers the government's ability to invest in new nuclear power, as well as impacting negatively on the public's perception of the industry.

The challenges associated with the clean-up of these wastes are vast, and difficulties with reprocessing are accentuated by a number of factors. The compositions of the sludges are highly heterogeneous and they are often flocculated, either from preliminary historical treatments or biological growths such as algae in open areas [1, 2]. Thus, they can compositionally change through transfer, especially in response high shear pumping. Also, there is a significant lack of characterisation data due to the high radiological and toxicological risks associated with sampling, and a large degree of variation in data from previous inventories. Because of these issues, there are many unknowns as to the potential behaviour of suspensions while they are pumped into separation corrals and interim storage tanks, before their eventual encapsulation. In particular, there is a defined engineering and scientific need to understand the settling behaviour of these complex sludges. The ability to measure concentration changes and stratification within these environments would greatly aid in developing efficient and cost effective separators, while using these data to enhance system modelling would, for the first time, give a truly predictive tool for process optimization. Indeed, the measurement and modelling of settling sludges is central to many multiphase engineering and environmental challenges, from water treatment to minerals processing, as well as in the oil and gas industry. In particular, the UK's water industry is a secondary market of concern, where considerable advancement in process performance must be achieved within the next five year cycle, due to rising energy and other costs in the regulated price market [3].

In response to these defined challenges, an important collaboration has been formed between the University of Leeds, Sellafield Ltd and MMI Engineering, as part of a £12 million pound R & D call into developing the civil nuclear supply chain led by the UK's Technology Strategy Board. Fundamentally, the project seeks to reduce the costs associated with sludge separation and increase process efficiency (thus reducing operational timelines) by developing two integrated characterization tools: 1) A predictive modelling framework; and, 2) An innovative and flexible *in situ* measurement technology.

The modelling tool is based upon computer codes that are presently used to predict the behaviour of clarifiers and thickeners in water treatment [4], and its extension for use in nuclear waste processing represents a major industry innovation. The framework combines models of sludge consolidation within a computational fluid dynamic (CFD) environment of separation systems, and has clear advantages in terms of safety, by reducing the necessity for onsite involvement. Presently, these methods are unfortunately only able to qualitatively predict nuclear systems due to the latter's complexity, and to better predict expected conditions, the model will be extended to allow for aggregate breakup and re-agglomeration (which is expected to be prevalent in the transfer of these wastes). Also, higher-order numerical simulation of particle deposition and agglomeration will also be used, enabling the refinement of the turbulence and particle modelling approximations used in the modelling tool.

Importantly, the modelling tool will also require verification from experimental data. To overcome the lack of sampling-based physical characterization, an *in situ* measurement device will be developed, with the ability to profile sludge density (particle concentration changes) in separation operations. Currently, there are no cheap and robust instruments capable of providing such information in a relatively non-intrusive fashion. Acoustic backscatter systems however *do* offer that possibility, and may potentially give multipoint depth profiles along with the flexibility of having a separated measurement array consisting of comparatively cheap and disposable transducers (where the main controller can be protected from the radioactive environment) [5-7]. Similar devices are currently used by sedimentologists to study particulate flows in coastal and river applications [8], but their use in radioactive sludge waste represents a significant technological advancement. The development of this acoustic characterization system will allow physical validation of modelling predictions, and give operators the ability to monitor changes during sludge separation in real-time, if required.

This paper reports on the initial collaboration outcomes, focused on the design of the acoustic backscatter array (ABA) and its controller. Also detailed is the design of a mixer-settler column required both to calibrate the acoustic transducers, as well as conduct the initial small-scale sludge settling trials. It is noted that research into the modelling package is not scheduled until 2014, to allow for the gathering of the physical comparison data, and hence will not be reported in this paper.

DESIGN & METHODOLOGY OVERVIEW

1. Design of the acoustic controller and acoustic backscatter array (ABA)

Although a number of commercial acoustic backscatter devices currently exist (for example; the Aquascat1000 from Aquatec Inc [9]) none have the flexibility to adequately measure the types of suspensions encountered in nuclear waste environments. Generally, these devices are used to study dilute sediment transport in environmental flows [8-10], and are not suited to concentrated heterogeneous sludges. The major restriction of commercial controllers is often their lack of configurability, as it is not often possible to accurately modify the transmitted signal specifications or refine the raw data analysis from the manufacturer’s standard settings. Hence, it is not possible to fine tune the controller for such difficult systems or run advanced excitation algorithms.

Due to these issues, a bespoke controller is being constructed based upon previous work by the Ultrasound Group at the University of Leeds, who have developed a highly configurable, multi-channel, acoustic controller called the Ultrasound Array Research Platform (UARP). The UARP is a 96 channel system capable of independently transmitting and receiving, in parallel, from multiple elements of the array transducer. Equally, the system can be configured to allow the connection of individual transducers via coaxial connectors. The development of the UARP has focused on designing a flexible platform that aids research. As a consequence, we are able to prototype new techniques that require individual element control and capture of pre-beamformed, raw RF data. The UARP consists of multiple 8 channel transmitter cards and 8 channel receiver cards connected via a backplane to a central field programmable gate array (FPGA) which acts as the central processing unit (CPU). The FPGA coordinates all transmitter channels and receives all raw RF data in parallel in real time. System control and raw RF ultrasound data are sent to a PC via a USB 2.0 link. Figure 1 illustrates the components of a typical UARP setup.

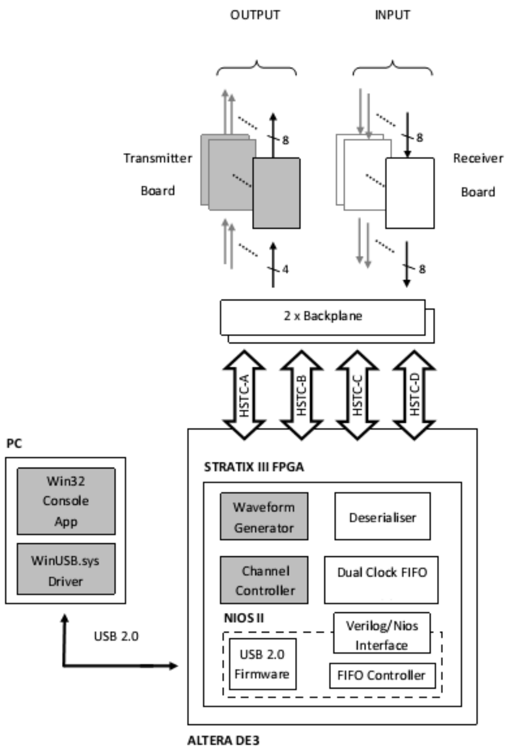

Figure 1: Schematic diagram of UARP V1.

Each transmitter card is fitted with an Altera Cyclone III FPGA and eight MAX4811 ultrasound pulser chips manufactured by Maxim Integrated Circuits. The Altera Cyclone III FPGA allows the transmitter to be locally driven in real time, reducing the design requirements for the DE3 main board. Additionally, the Cyclone III has been designed so that channel to channel time resolution control of <200 ps can be achieved [11]. Any tone or swept frequency excitation signal up to 15 MHz can be produced with the parameters controlled by a communications link to the DE3 main board. Options exist to use advanced excitation techniques developed by the Ultrasound Group to emulate arbitrary analogue excitation through the use of advance multilevel pulse width modulation (PWM) and to control harmonics through the use of Phase Inversion Selective Harmonic Elimination (PISHE) [12]. Combined, these techniques allow the control of excitation frequency, phase, amplitude and harmonic content. It is believed that this advanced control will allow new measurement techniques to be developed during this project.

The receiver card is fitted with a Texas Instruments AFE5805 ultrasound front end. The AFE5805 provides signal amplification and filtering plus analogue to digital conversion at 50 MS/s (million samples per second) and 12 bit precision on eight channels in a single chip. This level of integration and miniaturization is state of the art, allowing eight receiver channels to be fabricated with minimal components. To reduce the complexity of the AFE5805 the digital outputs are serial and operate at 600 MHz (12 bits multiplied by 50 MHz) and require a state of the art FPGA, like the Altera Stratix III on the main board, to decode and store the data.

A number of upgrades are envisioned to the baseline technology based on the UARP V1 ultrasound controller technology as shown in Table 1. It is planned to upgrade the AFE5805 to the newer AFE5807 allowing an increase in sampling rate from 50 MS/s to 80 MS/s. The communication system between the computer and acoustic controller will also be optimized and upgraded to an optional PCI Gen3 link. Additionally, the mechanical design of the RX and TX

PCBs will be changed to the *Eurocard* standard, offering improved mechanical stability and hence contributing toward the ruggedisation of the ultrasound controller for the planned field studies.

Table 1: Specifications for the Ultrasound Array Research Platform V1.0 and V2.0.

Specification	UARP V1.0	Proposed UARP V2.0
Number of TX/RX channels	8 channels per card 96 channels maximum	8 channels per card 64 per controller module 128 channels maximum
Transmit Frequency	up to 15 MHz	up to 15 MHz
ADC	AFE5805 (Max 50 MS/s)	AFE5807 (Max 80 MS/s)
Raw data transfer to computer	USB 2.0 (Max : 480 Mbps)	PCI Express Gen 3 (Max: 64 Gbps) USB 2.0 (Max : 480 Mbps)
Mechanical aspects	Wiring plus connectors on back-, side- and top-planes	Eurocard mechanical standard Connections routed via backplane

Another key aspect of the instrumentation is the design of the acoustic backscatter array (ABA). Acoustic systems commonly utilise probes in the 1 – 5 MHz range, and normally single transducers are mounted vertically down in the top of suspension columns or open flow estuary environments as, for example, in [8, 10]. Unfortunately, previous work by the current authors [5, 6] has indicated that for concentrated industrial sludges, acoustic penetration is much reduced in respects to their use in natural sediment studies. Therefore, to be able to observe behaviour along the full depth of large-scale settling systems, a number of transducers need to be mounted at different heights, and behaviour measured in segments. Therefore, a flexible and novel array holder was designed (as shown in Figure 2) giving the ability to hold up to 8 transducers on a single small-form vertical shaft made from durable Rexroth™ aluminium (of 1 m total length). The ABA uses 1 cm diameter piezoelectric ceramic probes, which can be held at various angles for flexibility. For the initial studies, the probes were positioned at an angle of 20º to the vertical. This angle was sufficient so that each probe pulse would not interfere or scatter off the transducers lower down in the array, while still being sufficiently close to vertical to be able to measure at depth. It is noted that the transducers are set with a mean 2.25 MHz frequency, although this can be varied by $^{+}$/- 10%. This flexibility gives the ability to obtain multiple frequency data from single probes, using variable excitation algorithms such as ‘chirps’.

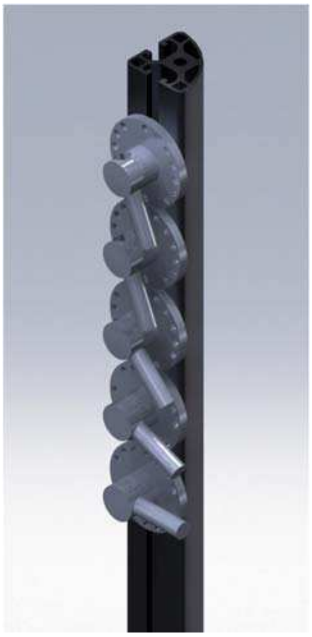

Figure 2: Design of the acoustic backscatter array (ABA).

For the first generation UARP system, the ABA can be separated by cables of up to 3 to 4 metres from the controller. However, for the full field instrument much longer cables (up to 20 – 50 m) may be required to fully isolate the controller and operator from the nuclear environments where the probes will be inserted. To achieve this extended configuration, signal booster amplifiers will be incorporated into connected cable sections to ensure sufficient transmitted voltage.

2. Calibration testing of the ABA in a mixer-settler column

Test material selection & batch-scale settling studies

To understand the performance of the ABA in measuring particulate segregation, different test materials were developed to replicate a range of behaviours common in nuclear waste sludges. It is noted however that at this stage, all testing performed was inactive, where the physical settling behaviour of the suspensions could be compared to a range of historical data on Sellafield wastes. Given in Table 2 is a list of the particle types, trade names and sizes for the 'as used' un-flocculated powders.

Table 2: Properties of test material powders.

Particle Type	Trade name (supplier)	Size, d_{50} (μm)
Spherical glass	Honite-16 (Guyson International)	41
Spherical glass	Honite-22 (Guyson International)	77
Calcium carbonate (calcite)	Omyacarb 2 (Omya Industries)	4.5
Magnesium hydroxide	Versamag (Akrochem)	3.4

Relatively monodisperse spherical glass powder in two size fractions was initially used as a well-defined sediment basis. The settleability of these suspensions is also comparatively easily predicted, giving an initial route to compare physical and modelling measurements of segregation. For more realistic test materials, it was decided to utilise flocculated fine particle systems, giving compositionally multi-component systems that mimic sludges found not only in nuclear environments, but across the minerals and water processing industries. Fine calcium carbonate (calcite) was selected, as a number of previous papers detail flocculated settling studies with this system as a model of minerals processing suspension [13, 14]. Magnesium hydroxide powder of similar fine particle size was also used. Magnesium hydroxide more closely mirrors the composition of corroded *Magnox* sludges that are common in Sellafield wastes [2]. However, as hydroxide dissociates from the particulates, such sludges buffer at high pH >10 – 11. Hence, there are potential issues with laboratory safety and instrument or rig wear under these conditions, and initial use of this material was therefore limited to batch-scale cylinder studies.

To flocculate the calcite and magnesium hydroxide, a number of commercial polyacrylamide type polyelectrolyte agents were sourced from SNF Ltd, as given in Table 3. For the calcite, a medium charge density high molecular weight anionic polymer was used (AN 934SH), as has been previously reported in the literature [14]. For the magnesium hydroxide, a high charge density version of the same co-polymer was utilized (AN 945SH), as it is better suited to high pH conditions. For comparison, a cationic polymer (FO 4650SH) was also used in flocculation studies, to observe the effect of charge type on performance.

Table 3: General properties of polymer flocculants used in settling studies.

Name	Charge type	Charge density	Molecular weight
AN 934SH	Anionic	Medium	High
AN 945SH	Anionic	High	High
FO 4650SH	Cationic	High	High

Design of the mixer-settler column

A critical component of the project, and the focus of our initial investigations, was to construct a large calibration mixing column, capable of generating homogeneous suspensions of the various test materials. Such a system is required to produce consistent particle concentrations at depth, and enable the calibration of the ABA transducers under repeatable conditions. Here, the profiled backscatter strength and signal attenuation can be measured over a range of different particulate levels, defining concentration correlations. This information in turn can be used to quantify density measurements from suspension settling studies conducted in the same column. Given in Figure 3 is a schematic of the column that also highlights the position of the mounted ABA on the front inner side (with the probes facing into the column).

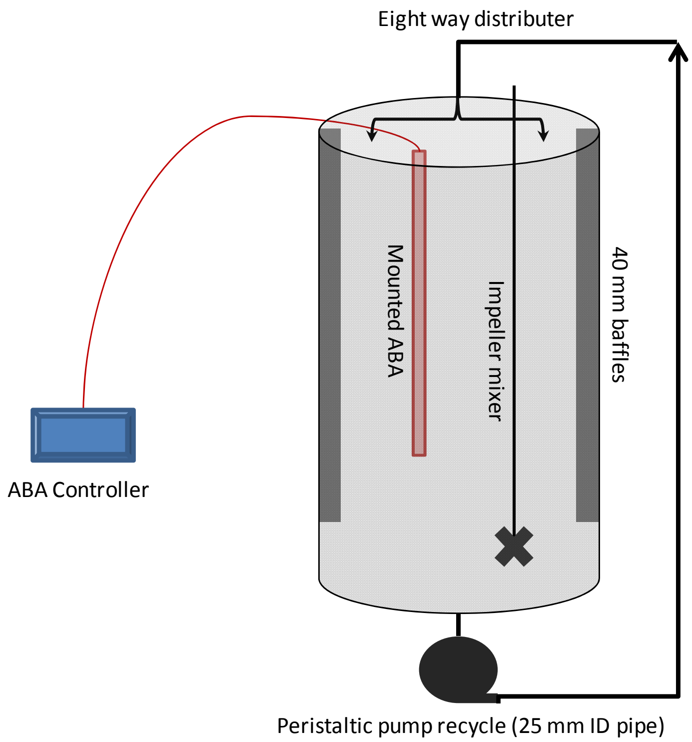

Figure 3: Schematic of mixer-settler column for ABA calibration.

The design of the column was complicated by two main factors. Firstly, it had to be large enough to allow for the acoustic spreading of the ABA pulses with depth, without the possibility of directional scattering off the walls or impeller that may lead to additional echo feedback. Considering a general spreading pattern of a standard 2 MHz transducer [15] with a half angle of 2.2^{o} (representing the lateral pulse width of half the acoustic energy) there is an estimated spread area diameter of ~7.7 cm at 1 m distance. Hence, a 0.5 m column diameter was used with an off-centre mixer (positioned 180 mm from the side) giving enough lateral area to position the probes as shown in the figure without the potential for internal interference. The column was designed with a 1.5 m total length (for a working testing depth of 1.33 m) and fill volume of ~261L.

Secondly, a relatively low-shear mixing system had to be employed to ensure minimal break-up of flocs under continual agitation. Hence, the column was designed with a small 100 mm axial flow impeller set to a low rotation speed using a variable inverter-mixer (from PH Pumps, Suffolk) and was used in combination with a peristaltic pump recycle (Watson Marlow 620s). The pump was set at a rate to approximately balance with the estimated settling flux of the suspensions, and the fluid was recycled by an eight way distributer back into the top of the column. Small 40 mm baffles were also constructed in the tank to reduce vortex formation.

Initially, trials were conducted with the un-flocculated glass particle suspensions. Sample measurements of concentration were taken to confirm the mixer and pump settings to attain homogeneous suspensions. Acoustic calibration experiments will also be performed with suspensions at various concentrations using the currently available commercial Aquascat1000 (from Aquatec Inc.) as a number of previous papers exist that detail the acoustic scattering and attenuation constants in these systems [5, 8, 10]. These results will be used to compare the performance of the bespoke ABA and controller in future studies planned for 2014. Scattering-attenuation correlations from flocculated calcite suspensions will then be compared, before a full set of settling studies will be completed in the calibration column, to obtain the required small-scale validation information for use in the modelling framework.

Preliminary column commissioning results

To firstly understand the quality of mixing using the off-centre stirrer and pump recycle, single phase tests were completed with NaCl solutions. Here, a concentrated salt solution layer (giving a density of ~1100 Kg/m^3) was made in the bottom 1/3 of the tank (to a height of ~50 cm). The tank was filled up with pure water above this mark (being careful not to disturb the dense salt layer). Then, the mixer and pump recycle were run for 15 minutes and conductivity measured to analyse the homogenisation. Given in Figure 4, are the average measured conductivities at different heights (in relation to the tank base) over time. It is clearly observed that complete mixing of the dense salt solution layer was achieved by around 5 minutes. For certainly, all tests with suspensions were firstly mixed for 15 minutes to ensure homogenisation.

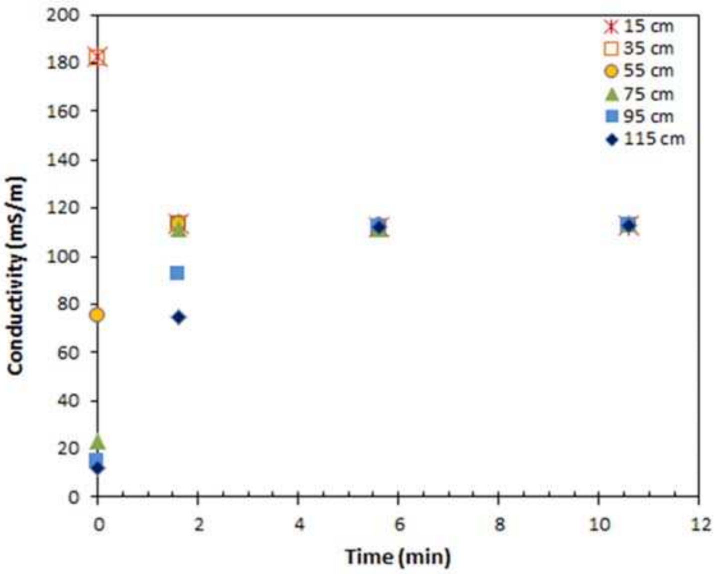

Figure 4: Conductivity of mixed salt solution layer in calibration column over time

To initially test the homogenisation capacity of the column with multiphase systems, Honite-22 glass particle dispersions (of 1 g/L nominal bulk concentration) were used, representing dense large particles of a size close to that expected from the flocculated test materials. Given in Figure 5 is the depthwise sampled concentrations, after mixing for 15 minutes.

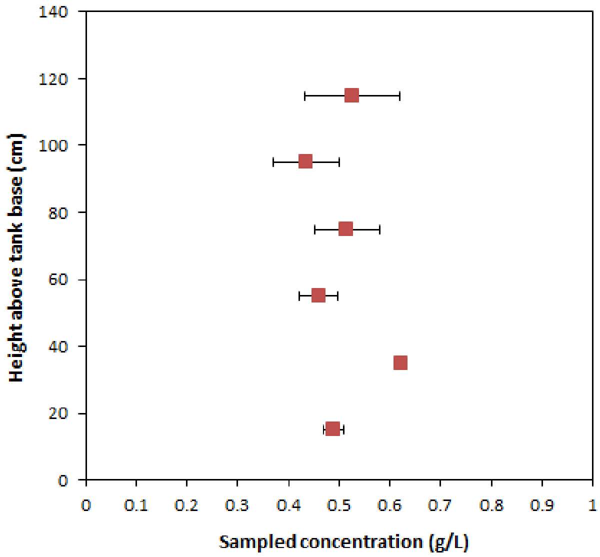

Figure 5: Sampled concentration profiles in calibration column with Honite-22

It appears from these data that although the combined impeller and pump recycle produces a relatively consistent depthwise profile (especially considering the sample variability due to the low particulate levels) all concentrations were below the nominal bulk value. The reason for this reduction was that the low shear impeller was not able to fully suspend particles off the base of the column at the sides (as could be observed visually). Hence, the larger fraction of particles was not being pulled through the pump recycled and remixed upon settling. We are currently modifying the rig to overcome these restrictions, by using a redesigned impeller which produces more uplift, and constructing a conical base insert to ensure all particles are pushed through into the recycle. It is again emphasised however that relatively low mixing shear is critical to help reduce breakup of flocculated systems, and importantly, it seems the inlet mixing in the top of the column from the recycle distributer is working effectively, as depthwise concentrations are relatively consistent.

CONCLUSIONS AND FUTURE WORK

Presented in this paper are the initial scoping studies and work methodology for an important industry-academic collaboration into developing innovative modelling and instrument technology to predict and measure nuclear waste transfer. Specifically here, the design of a flexible and powerful acoustic controller, the *Ultrasound Array Research Platform* (UARP) is detailed, which along with a novel vertical acoustic backscatter array, will allow for real-time *in situ* concentration profiling of sludge separation operations. Also described is the design and construction of a large mixer-settler column that has the capability to produce homogeneous flocculated suspensions with low-shear, which is being used both for the calibration of the ABA as well as conduction of the primary settling studies.

The ABA will enable suspension segregation to be investigated in a level of detail not possible in any current system. This in turn will give an unparalleled ability to verify new advanced drift-flux models, embedded in computational fluid dynamic models of sludge settling, which are also being developed as a future goal of the project. Indeed, although primary settling studies will be conducted in the mixer-settler column, future trials are planned with a ruggedized ‘site-ready’ ABA on a full scale non-active separation corral. This will for the first time give the ability to fully correlate and verify model predictions in a realistic and controllable environment.

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