Magnox Reprocessing TDN Reactors: Utilising 3D Printing and X-Ray Imaging to Re-Design and Test Fluidising Air Nozzles – 16402

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ABSTRACT

One of the key final steps in reprocessing spent nuclear fuel is the conversion of highly concentrated uranyl nitrate hexahydrate liquor [> 1000 gU/litre] into uranium trioxide (UO_3) powder by thermal decomposition using Thermal Denitration (TDN) reactors. The TDN reactors in the Magnesium non-oxidising (Magnox) fuel reprocessing plant are high aspect ratio fluidising beds, which suffer from poor fluidisation and UO_3 powder blockages. It is widely understood that these issues arise from the inadequate design of the fluidising air distributors (nozzles), which are located around the base of the TDN reactors. A non-active test rig was constructed at the National Nuclear Laboratory (NNL) experimental facility in Workington, Cumbria (UK) which used compressed air at ambient temperature to assess the performance of the current fluidising air nozzle designs (datum) against a range of new nozzle designs. Full-scale nozzle designs were manufactured using a 3-Dimensional (3D) printer and tested to obtain data of both quantitative and qualitative nature. NNL then collaborated with the Chemical Engineering Department at University College London (UCL) to further assess the fluidising potential of the new nozzle designs. Images of inside the fluidised bed were taken utilising a unique X-Ray facility. The results from this study will allow new nozzle designs to be recommended, manufactured and installed, which will ensure improved fluidisation of the TDN reactor bed and prevent blockages with UO_3 powder.

INTRODUCTION

Located on the Sellafield Ltd site, the Magnox reprocessing and uranium finishing plants have been the workhorse of the UK’s nuclear fuel reprocessing programme for over 50 years. The facility has taken spent fuel from across the globe and reprocessed over 50,000 tonnes into uranium trioxide (UO_3) powder for safe storage or to be re-used for conversion into reactor fuel in the future. One area highlighted for improvement was the conversion of uranyl nitrate liquor (UNL) into UO_3 powder; a process achieved through thermal denitration (TDN). This chemical reaction takes place in a fluidised bed reactor, which introduces hot fluidising air through nozzles and results in the liberation of large volumes of nitrogen oxides (NO_x), oxygen and moisture. This paper will describe how recent advancements in design of TDN reactor fluidising air nozzles have been achieved using test rigs at the National Nuclear Laboratory (NNL) and University College London (UCL), and discuss how and why 3D printing and X-ray imaging have been utilised to underpin the new designs.

Background

Once removed from a Magnox nuclear reactor, spent fuel is cooled for at least 90 days to allow short lived, highly radioactive isotopes to decay prior to the cladding being mechanically removed and the subsequent dissolution of the fuel in nitric acid. The uranium and plutonium are then separated from the aqueous solution through a process of solvent extraction. Once extracted, the uranyl nitrate solution is converted into UO3 powder using TDN fluidised bed reactors (Equation 1) [1].

Eqn. 1: $UO_2(NO_3)_2.6H_2O_{(aq)} \rightarrow UO3_{(s)} + NO_{(g)} + NO_{2(g)} + O_{2(g)} + 6H_2O_{(g)}$ $\Delta H = +586$ kJmol-1

It is generally accepted that the design of the fluidising air distributors, or nozzles, is one of the most important aspects influencing operation of the fluidised bed. The Magnox process TDN reactors have a high aspect ratio (they are tall and thin, see Figure 1) and contain 22 nozzles at the conical base; 21 smaller peripheral nozzles (Figure 2a) arranged in concentric rings and 1 larger central nozzle (Figure 2c). UNL is sprayed directly into the fluidised powder bed and UO_3 is produced when heat is supplied; driving forwards the highly endothermic reaction (Equation 1). The fluidising air is introduced at ~375 $^{\circ}$C and internal heaters are placed to ensure optimal temperatures are sustained within the bed. Additionally, external wall heaters act as a blanket to reduce heat loss.

In the past, studies have been conducted to assess the design and function of TDN fluidising air nozzles for Sellafield Ltd’s other reprocessing plant, the thermal oxide reprocessing plant (THORP), which commenced operations in 1994 THORP also uses TDN reactors for conversion of UNL to UO_3, however they have a higher aspect ratio compared with those in the Magnox process (Figure 1). Another difference is that THORP TDN reactors have only 4 smaller peripheral nozzles arranged in 1 concentric ring, and do not have a central nozzle.

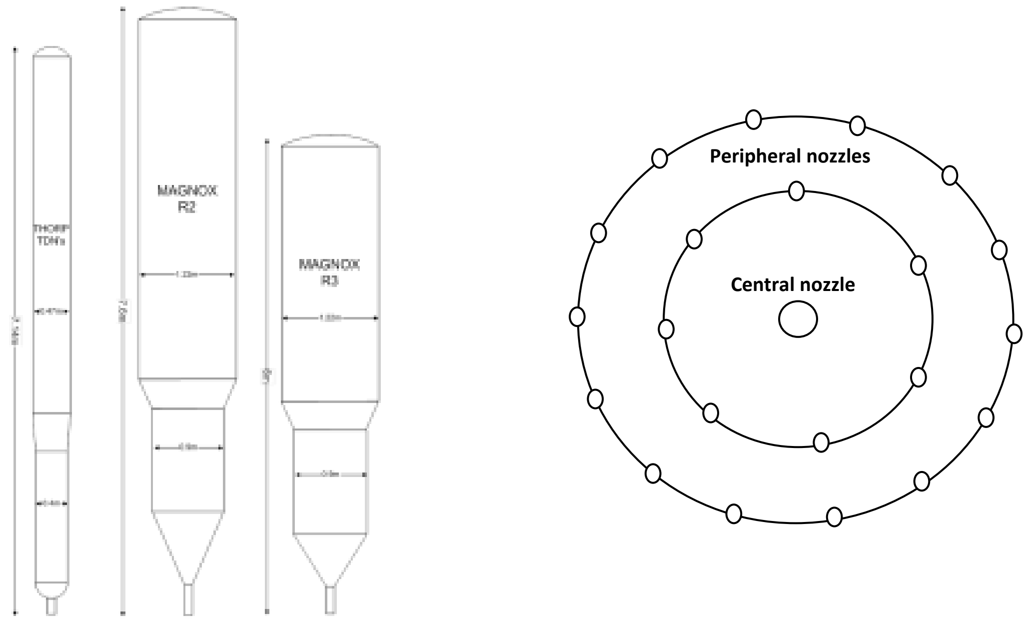

Figure 1: General schematic of Sellafield Ltd TDN reactors (left) [2], section of conical base (right).

In 1997, Geldart [3] proposed a new THORP TDN nozzle design consisting of three horizontal holes in the cylindrical nozzle wall. Old and new THORP nozzles can be seen in Figures 2a and 2b respectively. Installation of the new nozzle greatly improved TDN reactor performance in THORP, and has ensured continuous optimal performance ever since.

Figure 2: A selection of old and new nozzles, (a) Old THORP and current Magnox peripheral nozzle, (b) New THORP nozzle, (c) Current Magnox central nozzle

The current Magnox peripheral nozzles (Figure 2a) are almost identical in design to the pre-1997 THORP nozzles, and are thought to be responsible for a substantial portion of the difficulties that have been experienced in the operation of the Magnox process TDN reactors. When the fluidising air is switched off for maintenance there is considerable UO_3 powder ingress back through the nozzles and into the fluidising air inlet pipe. If there is moisture present, the UO_3 powder particles can bind to form a hard, ceramic-like material. As a result of this, the nozzles become blocked on reactor restart. Also, sub-standard fluidisation, caused by inadequate nozzle design, impacts on the heat transfer efficiency and results in formation of UO_3 clumps. This is due to the formation of a sticky 'mastic' intermediate phase, a stage which the chemical reaction can stall at if the fluidised bed is not sufficiently mixed. A further issue is that U_3O_8 (an undesirable product) can be formed at elevated temperatures. These temperatures can be achieved when UO_3 clumps form adjacent to internal heaters, then absorb and concentrate heat.

NNL Nozzle prototyping and testing

Building on the success of improvements made to the THORP TDN reactor nozzle design, Sellafield Ltd approached NNL with the task of investigating the design of Magnox process TDN reactor nozzles and proposing new designs that could alleviate the operational issues described above. NNL were heavily involved in the THORP nozzle redesign and the Magnox process project provided an opportunity for NNL to utilise their newly acquired 3D printing capability. 3D printing permitted multiple acrylonitrile butadiene styrene (ABS) plastic nozzle designs to be manufactured quickly, accurately and for a fraction of the cost and time it would take to produce metal nozzles. It is important to note that for the THORP nozzles, Geldart tested different designs using a single metal nozzle with both horizontal and vertical slits. Geldart then covered areas of these slits with tape to mimic different orifice sizes, shapes and geometries. Without the investment of significantly more time and money however, Geldart was limited to only the designs he could invent out of the single metal nozzle. 3D printing on the other hand, made it possible for a large number of different designs to be tested and fine-tuned as necessary, with the manufacturing process conducted for a fraction of the cost, in-house and completed in just a few hours.

EXPERIMENTAL

Test rig design

NNL built on site a fluidizing rig to adequately assess existing and new nozzle designs against a number of parameters. Photographs of the rig in various arrangements are shown in Figure 3. The main components of the rig were: pressurised air inlet, flow regulator, flow meter, flow controller, pressure relief valve, pressure transducer connections, nozzle, reactor column and simulant.
The air flows through the rig pipe work and out into the sand bed through a 12 mm internal diameter pipe that is internally threaded at the exit and contains two pressure transducer locations (Figure 3a). This allows different nozzle designs to be manufactured and screwed into the end of the pipe for testing. A plate that sits on the bench allows a 150 mm diameter Perspex column to be placed around the end of the pipe where the nozzle is attached, simulating the reactor bed (Figure 3b).

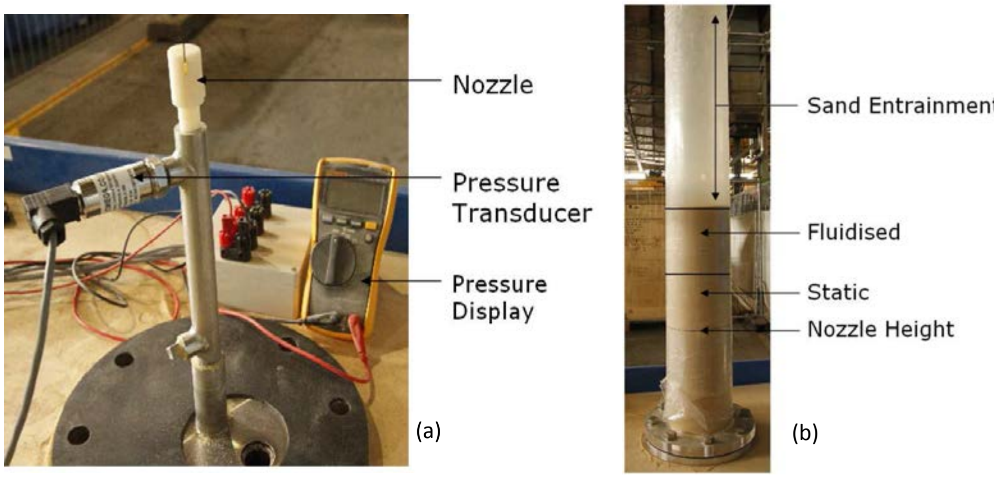

Figure 3: NNL rig arrangements [4, 5], (a) Pressure measurements, (b) Perspex column for fluidisation quality analysis

A larger Perspex column (400 mm internal diameter) was employed to test the central nozzle which normally processes higher air flowrates.
Kiln-dried sand (mean size 250 μm) was chosen as a simulant for the uranium trioxide powder of the TDN fluidised bed. This type of sand has been used in previous studies and is considered similar to UO_3 powder in terms of fluidisation behaviour.

The first parameter assessed was the discharge coefficient (CD), which is a measure of the nozzles ability to use the energy in the fluidising air efficiently. A low CD value means that the energy in the air is being lost within the nozzle itself, often due to turbulence or constriction of air flow. Conversely, an increase in CD means less air is used for a given amount of fluidisation work done, leading to potential cost savings. Since the purpose of the fluidising air is to mix the material within the reactor, it is desirable to ensure the energy held in the air is distributed into the fluidised bed, and not the nozzle itself. CD was determined using a pressure transducer, as seen in Figure 3a. Measuring the pressure in the pipe immediately before the air flowed through the nozzle allowed the pressure drop over the nozzle to be determined. This pressure measurement was used to calculate CD, and using the standard pressure drop over the Magnox TDN fluidised bed, the ratio of $\Delta P_{nozzle}/\Delta P_{fluidised\ bed}$ could be determined. The literature [6] reports that if this ratio was between 0.10 and 0.30, optimal fluidisation conditions could be achieved. Therefore, by optimising the design of the nozzle, a preferred CD value could be reached that would provide efficient and effective fluidisation.

In addition to quantitative analysis, the rig allowed a range of qualitative visual observations to be carried out. A Perspex column was secured over the nozzle outlet, simulating the TDN reactor. In order to assess some aspects of the fluidisation potential of each nozzle, the column was filled with sand (Figure 3b) and a filter placed over the top of the column. The parameters studied include the area of sand above the nozzle that remained static, termed “dead zone”, and the height to which the sand was entrained above the fluidised bed surface. In theory, the more effective nozzles would ensure smaller dead zones and lower sand entrainment values (at a given air flow rate), as this would indicate that the energy is being used to mix the bed rather than lift material into the air space above the powder bed.

The final measure was the ability of each nozzle to avoid becoming blocked with material. On completion of the fluidisation assessments, the air to the rig was stopped and the bed left to settle. A valve directly beneath the air inlet pipe was opened to measure the amount of sand that flowed back through the nozzle (simulating a blockage).

New nozzle design

The desired outcome of the preliminary experimental programme was to develop nozzle designs that provided more efficient and effective fluidisation, while being able to avoid blockages and mechanical deformation. The full range of larger central nozzles and a selection of smaller peripheral nozzles tested are shown in Figures 4a and 4b respectively.

(a)

(b)

Figure 4: A selection of 3D printed nozzles [4, 5], (a) Central nozzles, (b) Peripheral nozzles

Holes of different shape, size, angle, number and spatial arrangement were located on the nozzle wall as opposed to on the top face, in an attempt to eliminate powder ingress into the nozzle and prevent blockages. Some nozzles had holes on only one side of the nozzle wall, as these were designed to be installed in an area of the TDN reactor close to the reactor wall. This design consideration was to prevent air from being impinged on the reactor walls, which would likely result in erosion.

The results from the experimental programme were instrumental to understand why Magnox TDN reactors occasionally experienced operational issues. The visual analysis of the current nozzle designs confirmed the lack of mixing, as the fluidising air travelled straight up the middle of the powder bed, like a spout. This resulted in high levels and large quantities of entrained sand. The new nozzle designs generally mixed the powder bed much better, evidenced by varying magnitudes of dead zone decrease and lower levels of sand entrainment. The quantitative data also worked in favour of the new nozzle designs, as the $\Delta P_{nozzle}/\Delta P_{fluidised\ bed}$ ratios were well placed within the 0.10 to 0.30 optimal fluidisation range, whereas the current nozzles were at the very low end of this range. As for the discharge coefficient, it was discovered that as the $\Delta P_{nozzle}/\Delta P_{fluidised\ bed}$ ratio increased, the discharge coefficient decreased. Therefore a compromise was required in order to ensure optimal fluidisation, without significant detriment to the discharge coefficient.
The results from the preliminary study allowed a small selection of new nozzle designs to be taken to the next stage of investigation, which required a more in-depth study that would allow rigorously informed recommendations to be made.

X-Ray Imaging

Non-invasive experimental techniques are invaluable for providing a detailed insight into the flow patterns and general hydrodynamic characteristics of multiphase systems like fluidised beds. One such technique involves the use of X-rays and is based on the same principles as are used in diagnostic medicine to record the structures of body parts that are opaque to visible light.
X-ray imaging systems are composed of three elements: the X-ray source, the sample and the detection system (Figure 5). The sample interacts with X-ray beam, either absorbing or scattering X-rays, and the attenuated beam is detected in order to form an image of the sample. A unique high power pulsed X-ray generation system has been developed at UCL to provide X-ray pulses down to 200 µs width with an intensity of up to 450 mA at a voltage variable from 50 kV to 150 kV (Lettieri & Yates [7]).
This specially designed generation system has been successfully used [8] to study the hydrodynamic behaviour of a 4:10 scaled Perspex TDN reactor (Figure 5), operated on air and rutile following Glicksman scaling rules [9].

For this collaborative project, the non-intrusive nature of the X-rays gives confidence that the gas distribution and jet penetration lengths of the full scale nozzles can be accurately recorded, avoiding measurement artefacts seen with some alternative methods which can give misleading results. Representative air flow rates were obtained by matching the Froude number (the ratio of the flow inertia to the external field) between each compartment of the real Magnox process TDN and the cold model in sand.

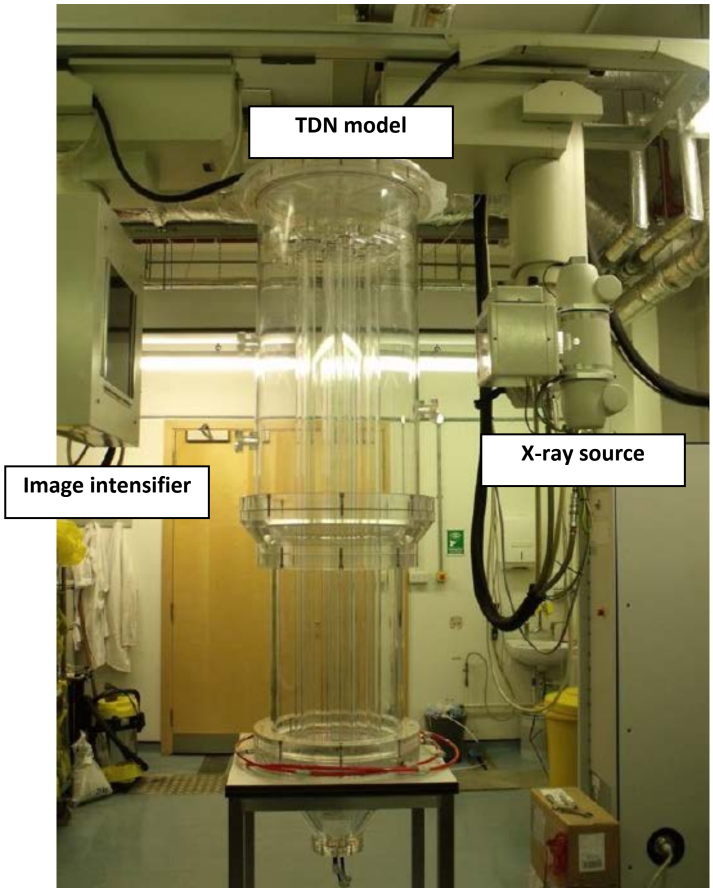

Figure 5: Perspex Model in the X-ray Cell at UCL [8]

RESULTS

Figure 6 below shows the X-ray image of tests conducted on the central nozzles at the same flow rates. The comparison was made between the proposed new nozzle design (Nozzle 26 - downward lateral holes) and the current central nozzles employed in the TDN Reactor 2 (Nozzle 18) and TDN Reactor 3 (Nozzle 28), both characterised by rectangular slots on top.

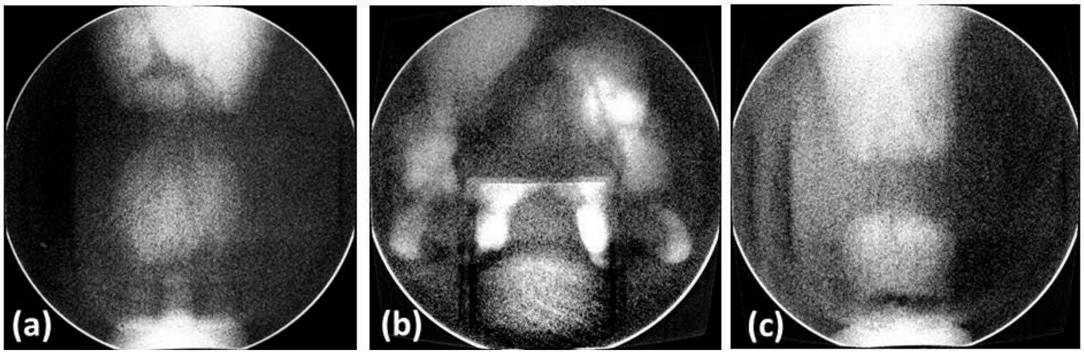

Figure 6: X-ray images in sand for central nozzles [10], (a) Nozzle 18, (b) Nozzle 26, and (c) Nozzle 28

Both current Magnox process central nozzles (Nozzles 18 and 28) produced a tube like pulsating flow with limited bed mixing, whereas the new central design, Nozzle 26, showed lateral penetration of the jets and the triangular pattern that the swirling gas produces as it converges above the nozzle. Coalescence of the jets exiting from the different holes is also clearly noticeable leading to greater gas distribution and solid mixing.

Different configurations were also studied for the smaller peripheral nozzles (Figure 7). Each peripheral nozzle processes significantly less gas than the central nozzle, but their function is of key importance in maintaining the fully mixed bed, thus enhancing the heat transfer with the internal tubular heaters. In the single slit configuration of the current Magnox peripheral nozzle design, the air plume appeared slightly unstable, wavering and pulsing under all flow conditions limiting the mixing to only the region above the nozzle. To remedy this limitation, new nozzles were designed with horizontally drilled exit holes distributed around all or half of the nozzle perimeter. This latter design was used as a nozzle directing gas into the center of the bed with the blank side facing outwards, thereby avoiding jet impingement on the reactor walls and any possibility of resulting erosion.

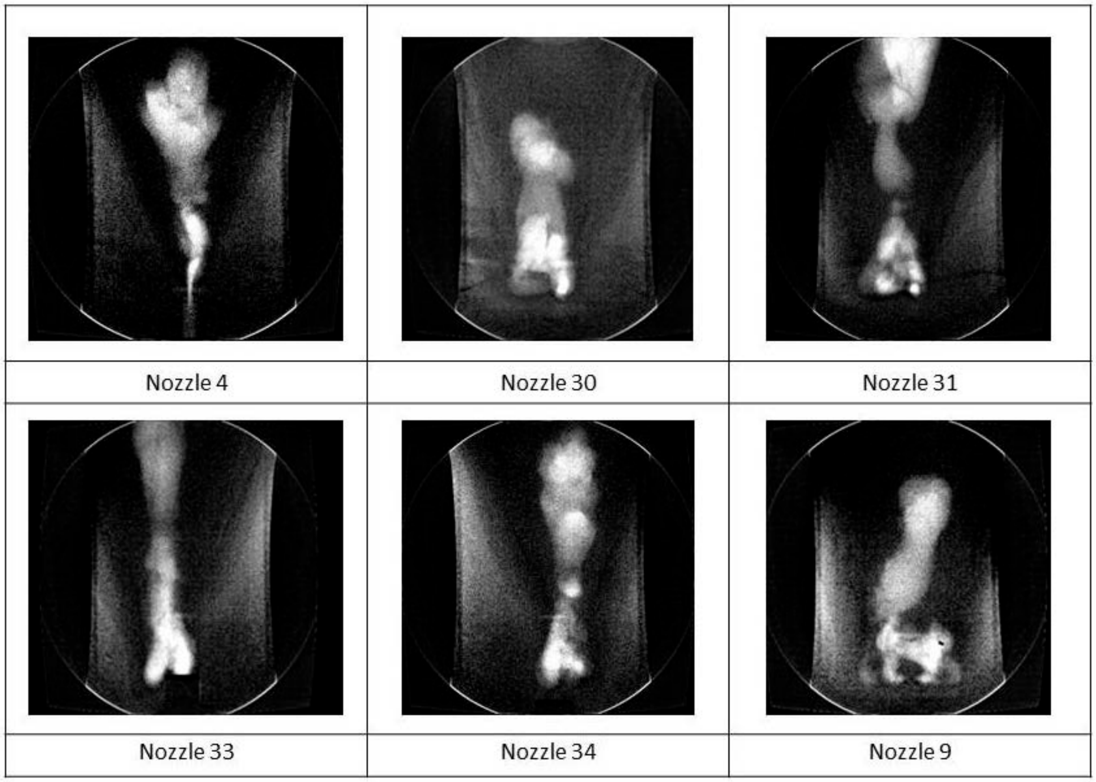

Figure 7: X-ray Images of jet penetration in sand at a 90 litres per minute flow rate [10]

As for the previous case, the images (see Figure 7) show a more stable plume/jet than in the slotted design (Nozzle 4) with a more even distribution and enhanced diffusion into the bed. The individual gas jets penetrate a few centimetres into the sand bed before merging above the nozzle about 5 cm above the gas jet inlets. Analogous behaviour was observed for peripheral nozzles with downwards angled holes (Nozzles 33 and 34). The downward pointing jet gives roughly half of the lateral penetration (~2 cm) compared to the horizontally directed jet (~3.7 cm). The average voidage profile could be obtained by averaging a set of images recorded in a certain time interval. Figure 8 shows the result of 720 images taken over a 10 second period. Nozzles with holes directed horizontally or at a downwards angle show a more even distribution of the gas and greater diffusion into the bed, with an average voidage distribution (y-axis) for the new design nozzles varying between 0.55 and 0.62, against 0.48 for Nozzle 4.

The most uniform distribution is obtained with Nozzle 9, designed with 6 horizontally drilled exit holes distributed around the nozzle perimeter. The design is the same as the one to be used as a central nozzle directing gas evenly into the bed, as shown in Figure 6b. The voidage profile of Nozzle 9, shown in Figure 8, shows the jets looping up and around, merging directly above the nozzle. The high voidage jets are well separated, as shown by the clearly visible yellow/green side jets.

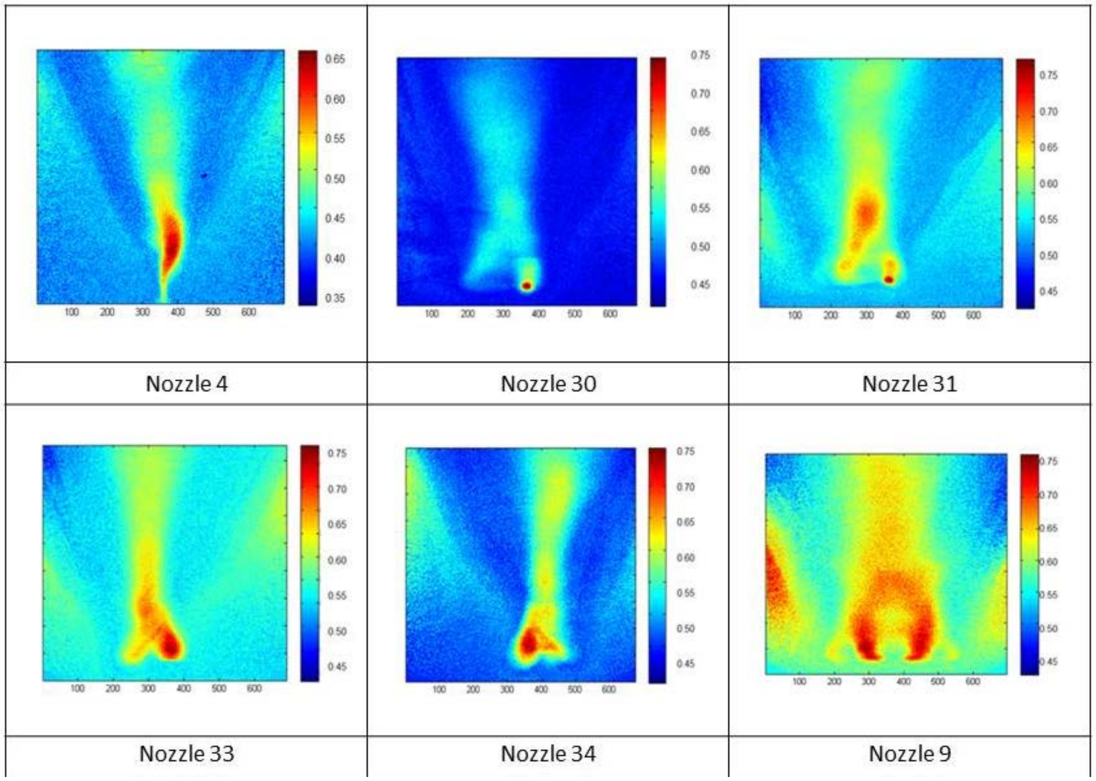

Figure 8: Average voidage profiles for peripheral nozzles in sand (720 images per nozzle) [10]

Figure 9 shows the jet penetrations as a function of orifice velocity. The jets all gave linear variation with velocity over the range studied, but the differences between the different designs are fairly small.

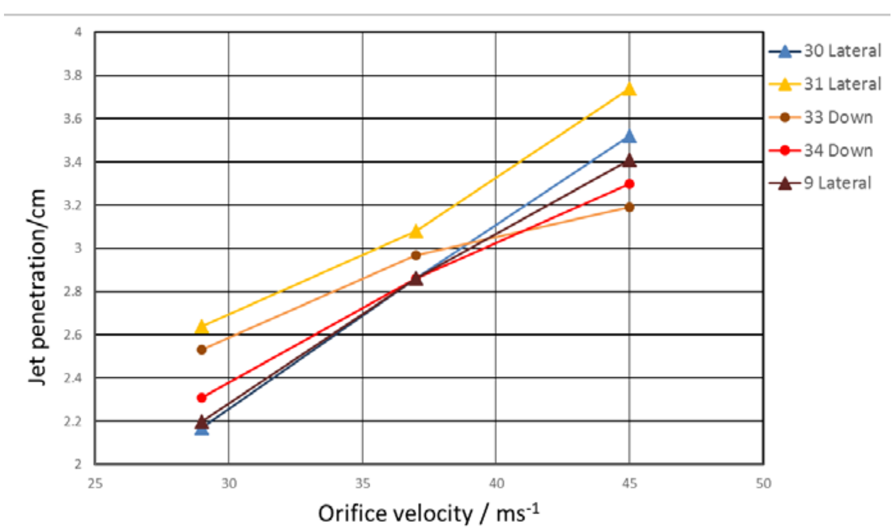

Figure 9: Jet penetrations v Flow Rate in sand bed [10]

CONCLUSIONS

This collaborative study into fluidising air nozzle design has contributed to a wider improvement programme (MTIP) aimed at establishing why the Magnox uranium finishing plant has experienced issues with poor fluidisation in its TDN reactors, and has provided a solution to this operational problem. From the NNL study it was found that a number of new nozzle designs with horizontally and downwards directed holes improved both the quantitative and qualitative parameters as compared with the current Magnox process TDN nozzles, which have rectangular slits directed vertically. UCL's study concluded that proposed new nozzle designs from the NNL study all result in improved performance over the current vertical slit design nozzles in terms of plume stability and gas distribution into the bed. Additionally, nozzles with downward pointing holes will clear the area below the jet outlets, but without any adverse effects on gas distribution, and should also help to minimise the tendency of the backflow of bed material into the gas distribution pipework. Finally, it was discovered that individual jets for all the multi-hole nozzles rapidly recombine a few centimetres above the nozzle. The new central and peripheral nozzle designs are due to be installed in the TDN reactors in 2016, and should help to ensure that Magnox reprocessing meets throughput targets with fewer operational issues than previously experienced. 3D printing has proven to be a quicker, more dynamic and cost effective method for manufacturing test pieces, and has massive potential to help not just in nuclear operations, but also in other areas of the sector, such as decommissioning and new build.

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ACRONYMS

ABS – Acrylonitrile Butadiene Styrene
CD – Discharge Coefficient
Magnox – Magnesium Non-Oxidising
MTIP - Magnox Throughput Improvement Plan
NOx – Nitrogen Oxides (NO and NO2)
NNL – National Nuclear Laboratory
TDN – Thermal Denitration
THORP – Thermal Oxide Reprocessing Plant

UCL – University College London
UNL – Uranyl Nitrate Liquor
ΔP_{nozzle} – Pressure Drop Over Nozzle
$\Delta P_{fluidised\ bed}$ – Pressure Drop Over Fluidised Bed

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