The development of centrifugal contactors

Next generation solvent extraction equipment for advanced reprocessing of nuclear fuels

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SUMMARY

- The technology gaps in our understanding of annular centrifugal contactors (ACCs) for the next generation of spent nuclear fuel recycling are being investigated in a modern setting
- This article reports on the unique-in-the-UK capabilities that the University of Leeds's Nuclear Engineering Group has established, as a partner of the AFCP, for testing and developing ACCs from laboratory to engineering scales and state-of-the-art computational fluid dynamics (CFD) models.

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1. INTRODUCTION

1.1 Introduction to the Advanced Fuel Cycle Programme (AFCP)

The UK has the challenging target to reach 'net zero' carbon emissions by 2050, as part of the global effort to mitigate the impacts of extreme, anthropogenic, climate change. The potential role that nuclear power can play in delivering this goal has been recognised in a recent White Paper [1] titled 'Powering our Net Zero future.' However, the trajectory for nuclear energy in the UK remains uncertain in terms of both the future capacity of nuclear to be installed and the types of reactors that will be built [2]. It also needs to be recognised that nuclear reactors are only part of the overall nuclear fuel cycle and require nuclear fuels for their operations and means of managing the fuel post-irradiation [3].

There are two basic nuclear fuel cycle options which are known as the 'open' and 'closed' nuclear fuel cycles respectively. Both cycles share the same operations up to the point that the fuel is discharged from a reactor. The decision is then to directly dispose of the fuel (open cycle or once-through cycle) or recycle and reuse the material as new fuel (closed cycle), as shown in Figure 1.

Previously, the UK has operated a closed cycle with reprocessing of spent fuel for recycling of uranium and plutonium. With the imminent end of reprocessing at the Sellafield site in Cumbria [4] current UK policy is to dispose of the remaining stockpile, as well as spent fuel from new build reactors in a geological disposal facility (GDF) from around 2080 [5] However, much can change over this time frame, and with no agreement on the site or size of a GDF, it is important to retain the option for spent fuel reprocessing, particularly if there is an expansion of nuclear energy within the UK and around the planet to help meet global climate change goals.

At a most basic level, recycling is more sustainable and more in line with the drive towards the 'circular economy' than disposal, as it reduces the needs for waste disposal, has lower environmental impacts, and makes the best use of natural resources [6, 7]. However, in the context of nuclear fuels, there are concerns over proliferation risks, economics, and technical maturity with closed fuel cycle option [8, 9].

As part of the Department for Business, Energy and Industrial Strategy's (BEIS) £505M Energy Innovation Programme (EIP), the £46M Advanced Fuel Cycle Programme (AFCP) was established in 2019 as a collaboration between BEIS and the UK's National Nuclear Laboratory (NNL). The AFCP represents the largest public investment in the research and development (R&D) of future fuels and fuel cycles in a generation.

Briefly, the AFCP aims to help the UK meet the net zero challenge by pioneering UK-made, globally deployed, next-generation nuclear fuels alongside salvaging valuable resources to increase the sustainability of nuclear energy whilst simultaneously minimising the environmental footprint. Sustainability, therefore, is at the heart of the AFCP ensuring it is recognised as a crucial enabler of the national net zero ambition, offering significant opportunities for the UK to reduce costs across future nuclear programmes, protect the environment, extend the lifetime of valuable materials and deliver huge economic benefit through driving environmentally-conscious, low-carbon energy innovation. The scope, structure, and aims of the AFCP have been described in detail in a previous article in this series [10]. Briefly, AFCP aims

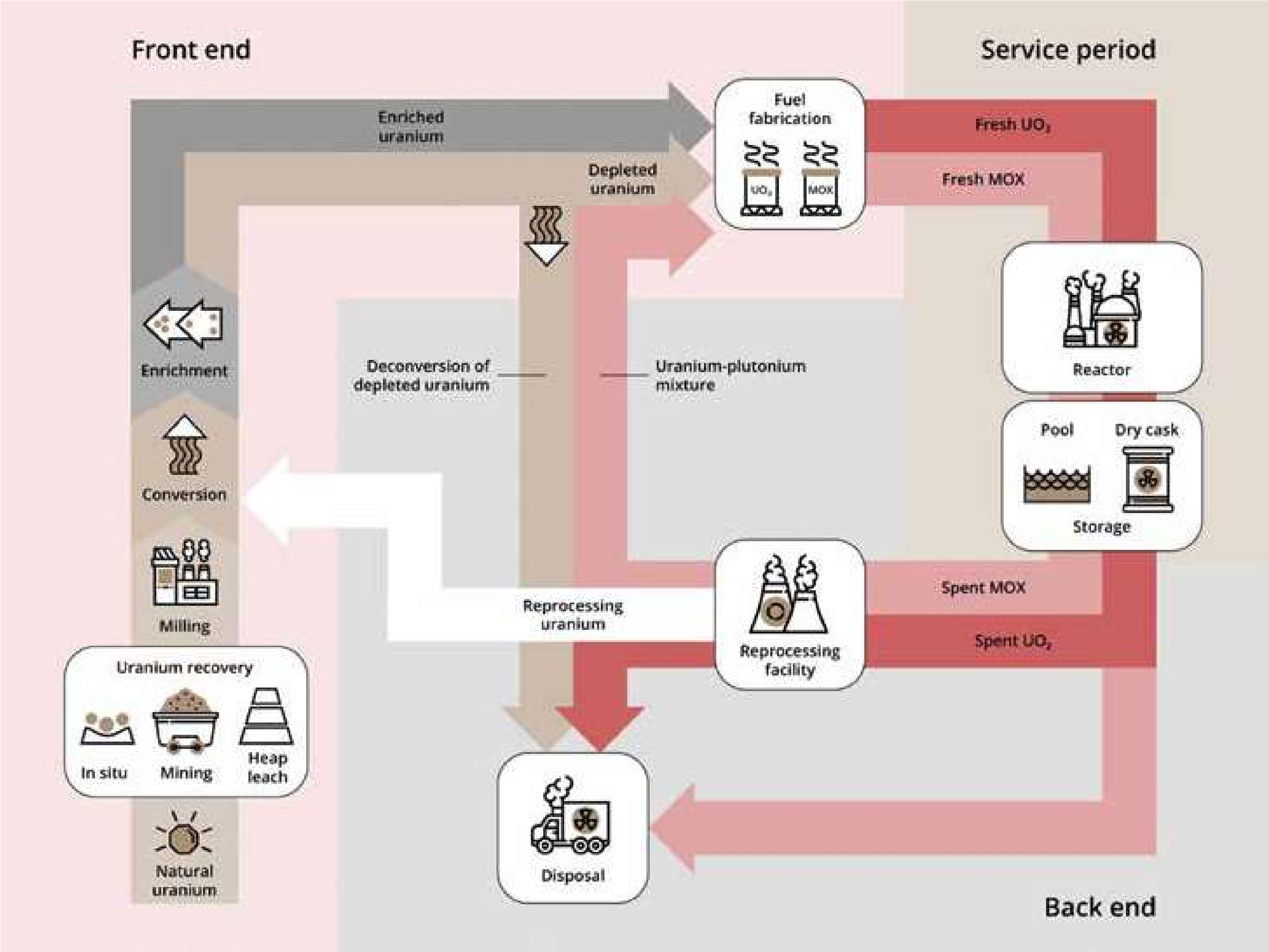


Figure 1: The nuclear fuel cycle from natural uranium through reactors and to ultimate disposal.

to develop advanced fuels needed for future reactors, such as advanced technology fuels (ATF) and coated particle fuels (CPF) in parallel with developing advanced technologies for recycling spent fuels – again see previous articles in this series [11, 12]

The objective for the 'recycle' projects within the AFCP is the development of proliferation-resistant advanced recycling options for spent nuclear fuels that generate less wastes, and are more compact and cost-effective, yet more flexible, than current reprocessing plants. Two basic strands of research are being pursued [13]:

- Advanced aqueous separation processes based on solvent extraction from nitric acid
- Pyro-chemical separation process based on electrochemical processing in molten salt media [14]

This research aligns the AFCP with international programmes and places the UK at the forefront of global research in this field [15]. The AFCP is designed as a collaborative programme and a wide range of academic and industry partners are working with NNL on both advanced fuels and fuel cycle research and technology development projects. One of the key collaborations within AFCP recycle project is between NNL and the University of Leeds on developing engineering scale demonstration rigs of various advanced technologies. New approaches that can reduce costs (through smaller footprints) reduce secondary wastes generated (through process intensification) and improve safety (geometrically safe units) are very attractive. One such technology is annular centrifugal contactors (ACC) for mixing and separation of liquids in nuclear fuel cycle separation processes and, whilst ACC have many intrinsic advantages, they have a low technology readiness level for recycle of irradiated nuclear fuel and significant R&D is still needed to underpin their deployment in a future fuel cycle plant [16].

Therefore, through the AFCP, the University of Leeds and NNL have established new capabilities for developing and testing ACC at scales from lab scale to pilot scale. This paper introduces ACC and then describes these capabilities and some of the results from the research at Leeds.

1.2 Background to reprocessing of nuclear fuel

The most common process used around the world for reprocessing nuclear fuel is the PUREX process. The PUREX process is based upon a chemical technique called solvent extraction, which is used widely in the mineral industry to purify metals.

Solvent extraction requires the metal to be in the form of a salt dissolved in an acid. This solution is contacted with an organic ligand (a specific molecule that has an affinity for the metal salt) and salt is transferred to the organic solvent from the acid allowing purification. The first step in any reprocessing plant is to get the fuel in the form of an acid solution, and this part of the plant is generally known as head-end. Once the fuel has been dissolved in nitric acid it is transferred to solvent extraction, this is referred to as chemical separation. The chemistry widely implemented is the Plutonium Uranium Reduction EXtraction (PUREX) process [17] and involves the use of the tri-butyl phosphate (TBP) ligand in an aliphatic diluent to extract uranium and plutonium from a nitric acid solution that contains all the soluble components of the spent nuclear fuel (SNF).

The PUREX process has been highly successful and implemented at commercial scales in the UK, France, and elsewhere [18, 19]. The PUREX solvent extraction processes are operated as counter-current processes, that is a series of interlinked solvent extraction devices that allow the solvent and aqueous to flow in opposite directions leading to quantitative extraction, stripping, and backwashing. Whilst the chemistry may be common across different facilities, the equipment used to achieve solvent extraction can vary. Typically, two types of equipment have been used in full-scale PUREX solvent extraction facilities, these being:

- Mixer-settlers
- Pulsed columns (also known as pulse sieve-plate extraction column or PSEC) [20]

Mixer-settlers (MS) consist of a mixing chamber into which immiscible solutions of solute and extractant are continuously pumped. The mixing chamber is a continuously stirred tank that generates a dispersion spilling over into a gravity settling chamber, where droplets of the two immiscible phases coalesce to allow separation. A review by Law and Todd from 2008 covers this equipment in greater detail [21].

PSECs consist of a column, into which immiscible solutions of solute and extractant are continuously pumped. The column has regularly spaced, perforated plates, through which droplets are formed. Mechanical energy is applied to pulse the column contents up and down, the pulsing action reduces the droplet size of the dispersed phase in order to improve mass transfer [21]. More information on these designs can again be found in the 2008 review by Law and Todd.

The current generation of reprocessing plants such as THORP (Sellafield, UK), UP2-800 and UP3 (La Hague, France), and Rokkasho-Mura (Japan) use PSECs and mixer-settlers [22]. There are many proven advantages to using these technologies such as reliability, low maintenance requirements, robustness to process variations, and tolerance of solid particles. Pulsed columns are also suitable for higher plutonium content streams due to their tall, thin geometry. Disadvantages of these technologies include:

- High capital cost
- In the case of PSECs, tall equipment leading to additional costs due to seismic requirements
- Longer residence times and higher hold-ups in the plant generating increased volumes of wastes from solvent degradation
- Mixer-settlers, of large geometric design, are less suitable for large-scale processes with feeds containing high fissile material contents
- PSECs are difficult to model and predict performance due to their dynamic behaviour

Therefore, as well as potential improvements to the *chemistry* of the PUREX process (or development of other more innovative separation processes based on solvent extraction, such as GANEX or Grouped Actinide Extraction) [23]. Improvements to *chemical engineering* should also be considered to address these shortcomings. Ideally, we would like technologies that can be designed to be geometrically safe, have low hold-ups, short residence times with intensified mixing and settling, are low and compact, and are able to be modelled.

The next generation of solvent extraction technology is being widely considered as a successor for MS and PSEC, known as the annular centrifugal contactors (ACCs), also known as centrifugal extractors, annular centrifugal extractors (ACEs), or simply centrifugal contactors (CCs). ACCs are more space efficient and of more compact design compared to the existing MS and PSEC [24], but have not been implemented in a UK nuclear reprocessing plant, so there is a great deal of knowledge required to determine their feasibility. However, they do have significant advantages including:

- Intensified mixing and settling in very compact units
- Low profile (unlike PSEC) and small size (unlike MS)
- Short residence times mean reduced solvent degradation from radiolysis and acid hydrolysis, as well as reduced hold-up in the plant
- Critically safe geometries

1.3 Review of Annular Centrifugal Contactors and their use in reprocessing

ACCs consist of two regions; a mixing zone (the annulus) and a separating zone (a rotating bowl), as shown in Figure 2.

The acid feed (with the SNF components within it) is fed into the annulus at the same time as the organic feed (consisting of the tributyl phosphate (TBP) in diluent). Within the mixing zone, the rotation of the bowl causes high shear which creates a dispersion of droplets with a high surface to volume ratio which is ideal for high rates of mass transfer. The dispersion enters the separating zone, where the two liquid phases are separated using centrifugal force, as they are immiscible. The advantage of using centrifugal force is that it can be up to 200 times greater than gravity which is the main separation force for MSs and PSECs.

ACCs are principally scaled-up by increasing their rotor diameter (RD). A typical rule of thumb is that 1-25 mm RD is defined as lab-scale, 25-51 mm is pilot-plant scale, and greater than 51 mm would be full scale. However, ACCs come with disadvantages such as increased engineering complexity, susceptibility to blockages by solids, unproven reliability, and higher maintenance requirements [26].

That said, ACCs have been deployed in the nuclear industry on a plant-scale [27], where the timeline of deployment is shown in Figure 3. Following the initial development by Webster and co-workers, in 1963, of an ACC at the Savannah River Plant [28], modifications were made by Bernstein and co-workers at Argonne National Lab (ANL) where they replaced a paddle mixer with fixed baffles at the base of the mixing zone [29-32] to

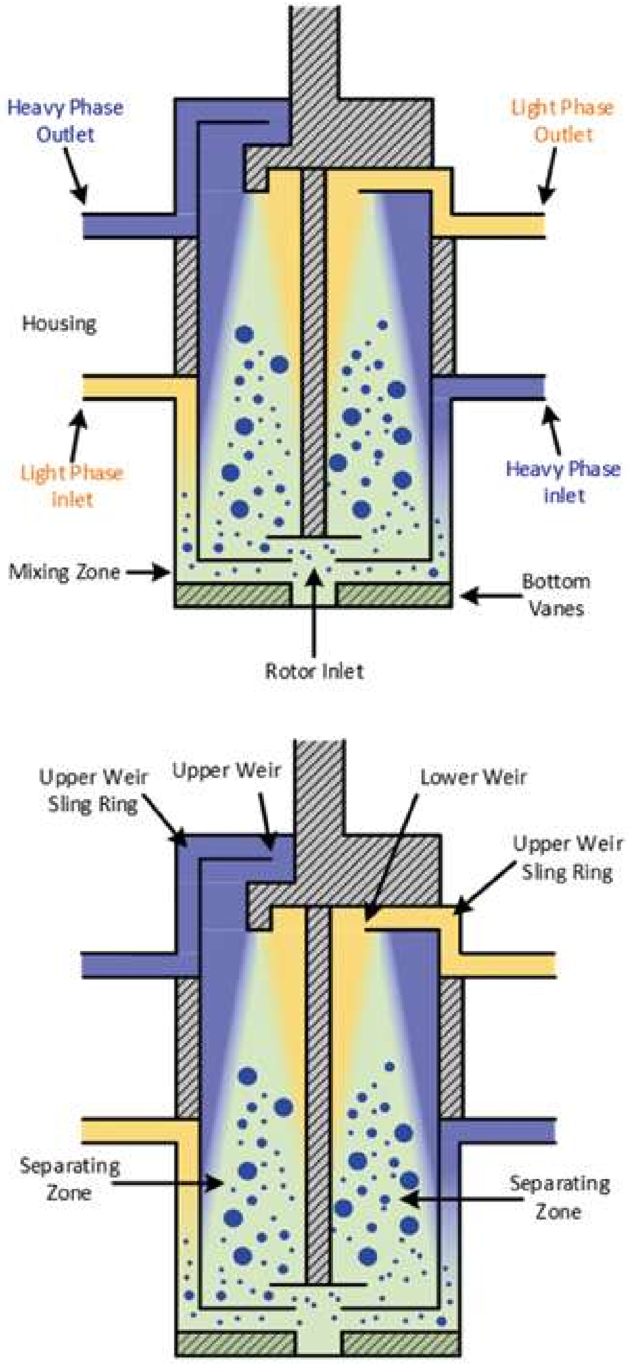


Figure 2: Centrifugal contactor diagram [25].

produce the design of an ACC that is now the basis of those that are commercially available. Since 1963, there have been four examples of ACCs on reprocessing plants [27].

The earliest reported deployment of ACCs was at the Savannah River Site (SRS) in 1973 [28] specifically in H-Canyon which was one of two reprocessing production lines [33]. This plant used a cascade of 18 stages of 250 mm RD ACCs for the chemical separation, based upon a PUREX flowsheet. Blocks of 6 ACCs were grouped together inaptly named '6-packs.' These were designed so that they could be interchanged rapidly for maintenance/repair or disposal [27].

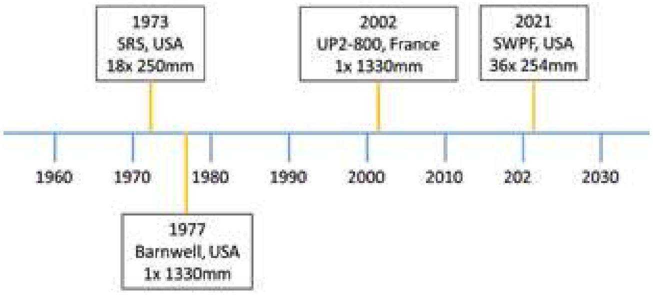


Figure 3: Timeline of industrial deployments of ACC in nuclear fuel reprocessing around the world [27].

In 1977, the Barnwell Reprocessing plant began cold testing of a single 1330 mm RD multistage centrifugal contactor [26]. This design is different to previous ACCs as a single unit contains multiple mixing and separating zones as shown in Figure 4.

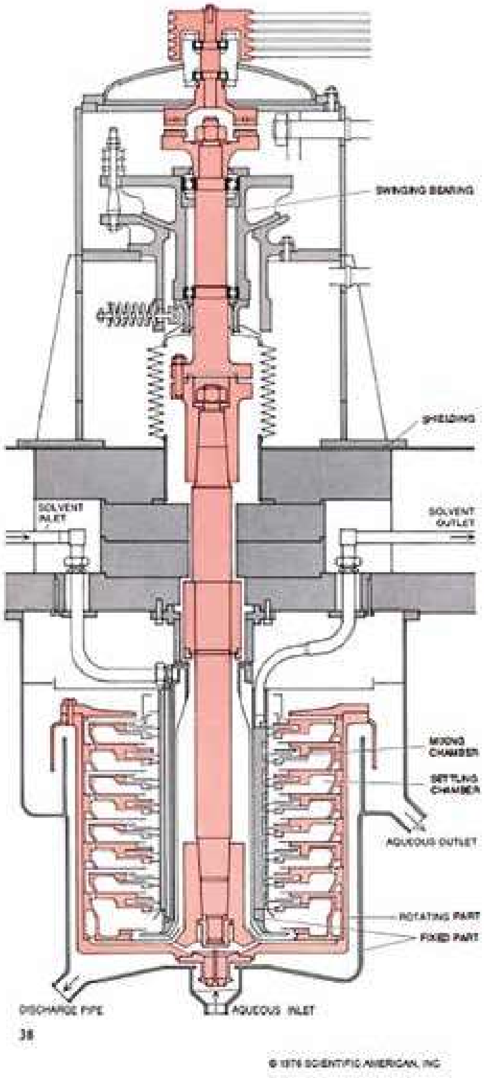


Figure 4: Multistage centrifugal contactor [34].

This multistage centrifugal contactor was to be used in the first cycle of highly active extraction of uranium and plutonium. Master-Slave Manipulators were in place to facilitate removing the multistage centrifugal contactor and placing it into a shield carrier to a suitable repair or disposal station.

Figure 5 shows the location of the multistage centrifugal contactor within a cell at Barnwell. Although the cold testing was successful, due to a change in the political climate, the plant never saw 'hot' commercial operations.

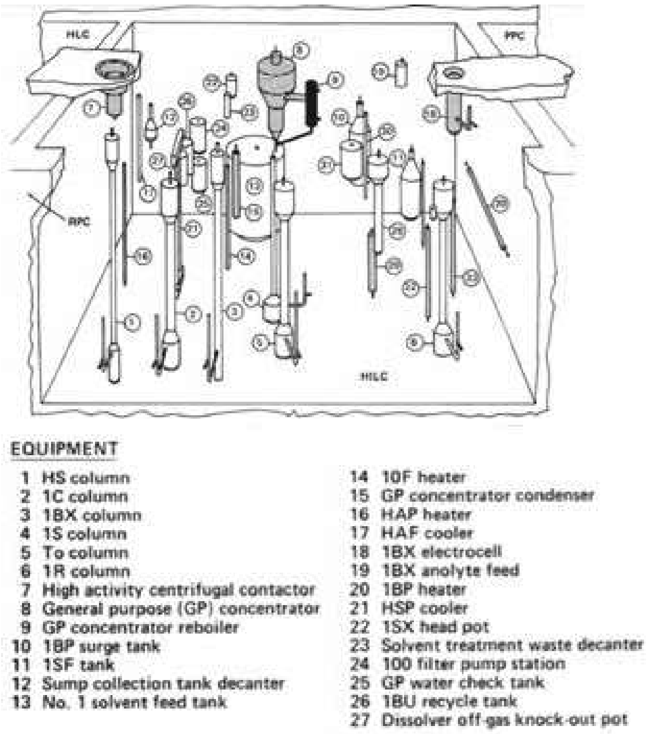


Figure 5: Processing equipment associated with PUREX co-decontamination plant in the high-intermediate cell components at Barnwell plant [35].

Another multistage centrifugal contactors was installed in 2002 at the UP2-800 reprocessing plant in France for the plutonium purification, and has been operated without a reported incident [36, 37]. More recently in 2021, there were reports that the Salt Waste Processing Facility was commissioning a cascade of 36 stages of 254 mm RD ACCs for the removal of radioactive isotopes cesium-137, strontium-90, and actinides. Table 1 summarises the limited deployments of ACCs [27].

There is, therefore, limited industrial experience in using ACCs and particularly the modern stage-wise designs [38] although commercial ACCs from suppliers such as Rousselet Robatel are publicised in deployments for pharmaceutical manufacturing, purification of polymers and food products, parachemical industry and minerals and mining. Deployment of ACCs in a nuclear environment is unproven in the UK and so there is still a need to raise the technology readiness level.

An example of demonstrating the engineering challenge and the ability of ACCs to scale up can be found in Japan. The Japanese Atomic Energy Agency (JAEA) constructed the Advanced Centrifugal Contactor Test System (ACT) which is an engineering test facility to build upon their small-scale ACC experience [27]. The ACT consisted of a three-story structure with the ACC cascade located on the second floor, shown in Figure 6. PUREX trials simulating throughputs of 10 kg of uranium per hour were performed in 2009 on the cascade of 34 stages of 80 mm RD JAEA design ACCs with remote maintenance. Mal-operation testing was performed alongside testing advanced ACCs of internal circulation type with magnetic bearings. No issues of solid precipitation or third phase formation arising were reported [39, 40].

The use of laboratory scale ACCs is shown on the timeline in Figure 7. The earliest use of ACCs in the UK open literature was in 1993 AEA-Technology Ltd in Harwell and Culham.[27] Three blocks of stainless steel containing four 55 mm RD ACC of Oak Ridge National Laboratory design were acquired and tested. These blocks were combined to form a cascade of 12 stages and used to test PUREX flowsheets [41]. Testing was continued at the British Nuclear Fuels plc during the 1990s with both 55 mm and 100 mm stages with limited success [42].

Then for more than two decades, NNL has used laboratory-scale ACCs (10 mm RD supplied by the Institute of Nuclear and New Energy Technology (INET), in China) [27]. Originally established by BNFL in 1996 before being transferred to the new PuMA Lab in NNL's Central Laboratory complex at Sellafield in 2010, NNL has multi-stage cascades for testing flowsheets and single stages for mass transfer studies housed in both glove box and fume hood for plutonium and low alpha activity (uranium) based tests [23, 38, 43, 44]. The use of ACCs in this context is to confirm that the flowsheet chemistry is compatible with the very short residence times for fluids in ACCs, rather than to test the ACC technology per

Facility	Process	ACC	Aqueous throughput (L/h)
F-canyon PUREX, Savannah River Site	PUREX HA first cycle, and second Pu cycle	18x 250mm, Unknown manufacturer	unknown
Barnwell Nuclear Fuel plant	PUREX HA first cycle	1x 1330mm* RR LX 670 N	3500a
R4 UP2-800, Cap la Hague Reprocessing Plant	Plutonium purification	4x 1330mm* RR LXN design	3500a
Salt Waste Processing Facility, Savannah River Site	Cs removal, Caustic-Side Solvent Extraction	36x 254mm, CINC V-10	3420

Table 1. Reprocessing/ waste processing plant deployments of centrifugal contactors [27]
***assumed, a minimum reported by Rousselet Robatel.**

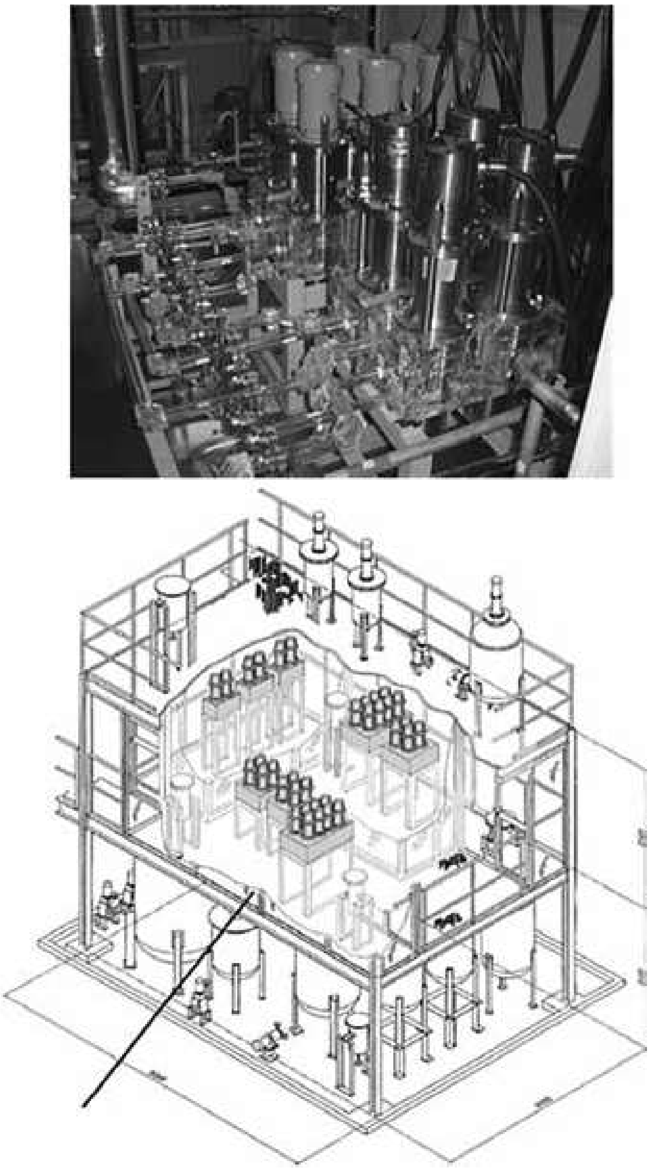


Figure 6: Top- JAEA designed ACCs of 25mm RD. bottom-Advanced Centrifugal Contactor Test System (ACT) installed at JAEA [39, 40].

se. Example results of flowsheet tests performed at NNL in ACC cascades are described in a range of publications [43-45].

As well as the hydrodynamic behaviour and defining suitable operating envelopes, perhaps the most important feature of the extraction process is to understand how mass transfer varies between the scales from lab to pilot to industrial scale. However, there is only one experimental evaluation across scales and this work did not look at mass transfer specifically [46]. In a review by Vedantam and Joshi in 2006 of the entire body of work on ACCs, it is stated that "relatively scant information is available on the detailed quantitative flow pattern in single and multiphase flows, mixing and axial mixing, mass and heat transfer coefficients, and drop and bubble size distribution" [47].

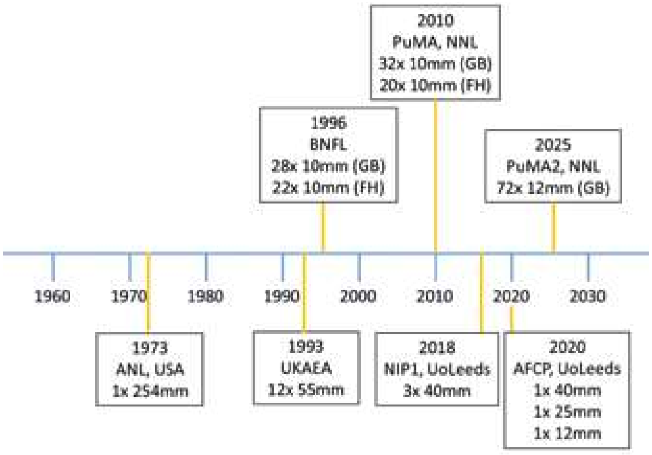


Figure 7: Timeline of UK Nuclear deployments of ACC in nuclear fuel reprocessing; GB= Glove box, FH=Fume hood [27].

NNL is currently looking to expand the capabilities for flowsheet testing and isotope recovery with a new laboratory (PuMA-2) of a cascade of 72 stages of 12 mm RD ACC, which is expected to be operational around 2025. However, on this timeline, the UK has only deployments of single sizes of ACC, and as a result, there is a gap in physical understanding of the effects of scaling to different sizes from the miniature ACCs. This is the scope that the University of Leeds's Nuclear Engineering Group, as a partner of AFCP, has been tasked with filling, both through physical experimentation and digital modelling. Specifically, investigating the effect of scale-up and technology design on the performance of ACC up to the engineering scale. As part of this work, the impact of solids upon ACC performance is a key factor to understand the suitability of ACCs for use in the first solvent extraction/scrub stages, which may contain fine particulates after clarification.

In 2020, the University of Leeds commissioned a physical rig of consisting of three different RD ACCs, and simultaneously began developing computer models of ACCs. The ultimate goal is to provide a digital twin that is validated through physical experiments.

2. ANNULAR CENTRIFUGAL CONTACTOR DEVELOPMENT IN THE ADVANCED FUEL CYCLE PROGRAMME

Innovation Programme (NIP, 2017-2019). In this work, a pilot-plant scale rig was developed for investigating a cascade of 3 stages of 40 mm RD ACCs to examine the effect upon performance of suspended solids [13]. However, the issue of scale-up (from 10 mm RD INET ACCs to pilot-plant ACC diameters and beyond) was still needed to be evaluated, in order to predict the performance at full scale.

Figure 8 pictorially sets out the avenues of ACC investigations at the University of Leeds, focusing on the effect of scale-up on hydrodynamics and solvent extraction, as well as the influence of particle slurries in a cascade of ACCs. Understanding both of these objectives over the past four years has been crucial to informing future computational models of the total throughputs and, therefore, the design requirements for future reprocessing technologies.

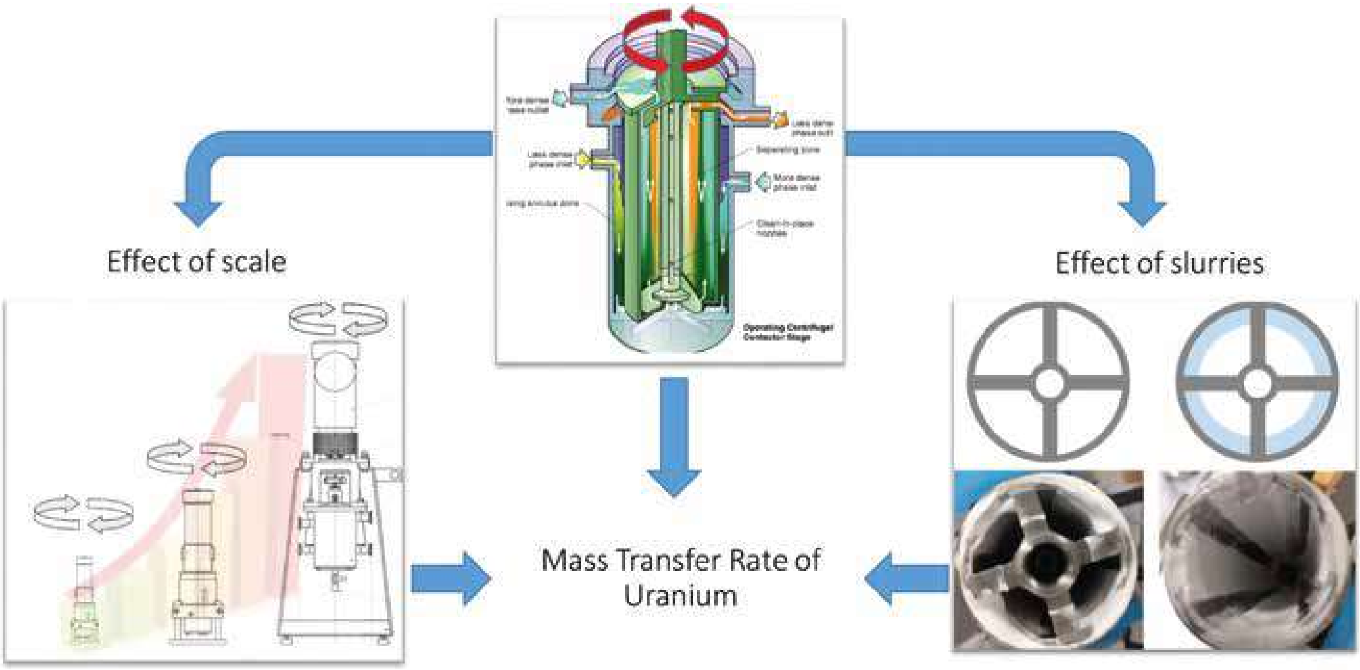


Figure 8: University of Leeds Experimental program.

Therefore, in the second stage of AFCP, the University of Leeds constructed a multiscale contactor rig (MSCR) for performing experiments addressing knowledge gaps in fundamental scientific understanding and plant operability for the industrial development of ACCs. Incorporated was a new experimental rig with three sizes of contactor that was designed and built (12 mm, 25 mm, and 40 m diameter) using commercial off-the-shelf (COTE) ACCs, so that scale down (to laboratory size from the Phase 1 3-stage pilot-plant rig) could be assessed; this is shown in Figure 9.

The MSCR has been built so that the auxiliary equipment and instruments can be used across the multiple scales with a simple turn of a valve. The MSCR encompasses online process analytical technology in a uranium-active setting, which included temperature, pressure, and UV/vis spectroscopy.

A sketch of the MSCR is shown in Figure 10 top left which shows the proposed arrangement of the non-active feed tank, pump, ACCs, and control units. Figure 10 top right shows the four tanks at the back; one aqueous feed tank, one solvent feed tank, and two receiver tanks for the outlet streams.

The first set of experiments was a series of hydrodynamic studies using aqueous nitric acid and TBP/OK, to determine the rotor speeds that allow efficient solvent/aqueous separation. Results are shown in Figure 11. From the initial series of experiments, the minimum rotor speed for efficient solvent/

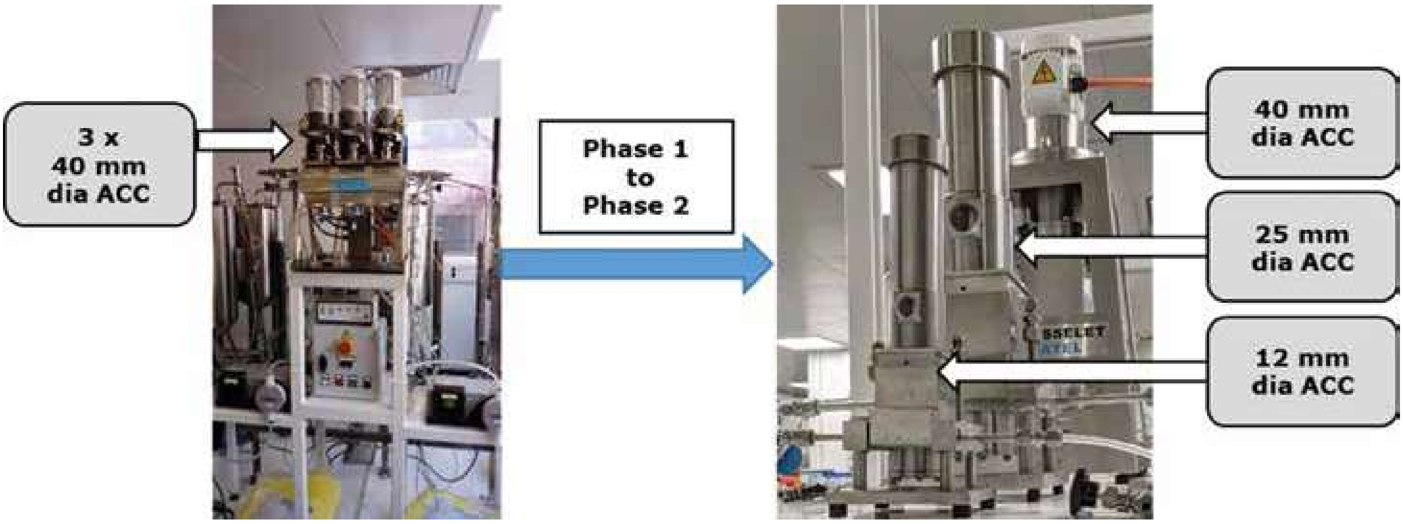


Figure 9: Scaling of centrifugal contactors.

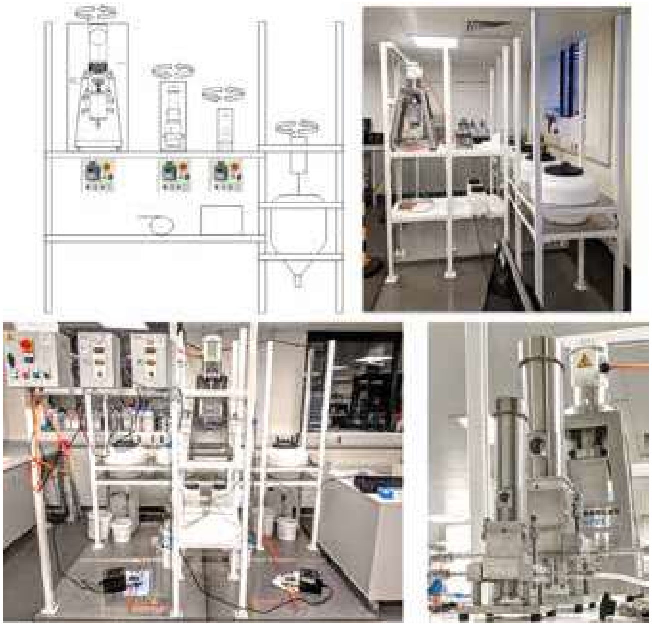


Figure 10: Multi-scale centrifugal contactor rig: top left – sketch of MSCR, top right – side view of MSCR bottom left= front view of MSCR , bottom right=close up of three sizes of ACC.

aqueous separation was determined. Figure 11 shows that 100% aqueous (Aq.) entrainment occurs as the rotor speeds are reduced. At these rotor speeds, centrifugal force is insufficient to overcome the gravitational force, resulting in flooding, and only flow from the solvent outlet is observed as it is lower than the aqueous outlet, and gravity follows the path of least resistance through the lower weir separator.

Upon determining the regions of hydrodynamic stability on the MSCR, solvent extraction studies with uranium were performed. Figure 12 shows the result of a trial on the 25 mm RD ACC using 10 g/L aqueous uranium. The ACC was run at a speed of 3500 RPM and fed with a total throughput of 60 mL/min, with a S/A ratio of 1. In this experiment, the ACC was fed with non-active feeds for 4 minutes to allow it to fill. Then, the uranium aqueous feed was started and this was defined as time zero in Figure 12. After 15 minutes of aqueous feed, it was switched back to non-active feed for the final 5 minutes to run down. The change in feed concentration is shown as the red line in Figure 12. Steady-state appears to be reached after 5 minutes, the mass balance was within ±5% which is generally considered acceptable, but the stage efficiency is <10%. During the run-down, it is evident that uranium is still present in both solvent and aqueous outlet streams, which gives an indication of the holdup within the ACC and its residence time. Further (ongoing) investigations, for example using dye injection, may be able to determine actual residence time experiments.

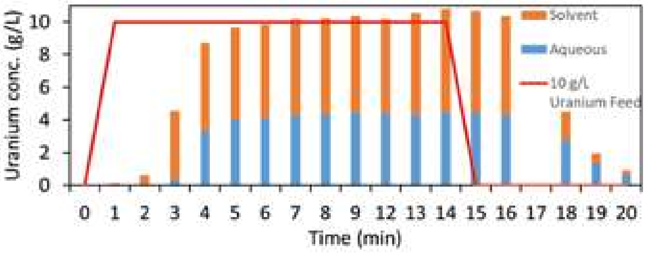


Figure 12: Solvent extraction of aqueous uranium on the 25 mm RD ACC with a rotor speed of 3500 RPM and a total throughput of 60 mL/min and a S/A ratio of 1. The redline shows the feed concentration of uranium (10 g/L).

Studies at the University of Leeds have also investigated the tolerance of ACC to solid particles in the aqueous feeds. The University of Leeds commissioned a centrifugal contactor cascade rig that consisted of a cascade of 3 stages of 40 mm RD ACCs, shown in Figure 13.

Solid aluminium oxide particles were suspended in the nitric acid aqueous feed and pumped into the cascade. It was discovered that the intense shear mixing in the mixing zone of the ACC maintained the suspension of solid particles; however, the intense separator G-force resulted in a thin sediment layer formation. The majority of solids accumulated in the feed

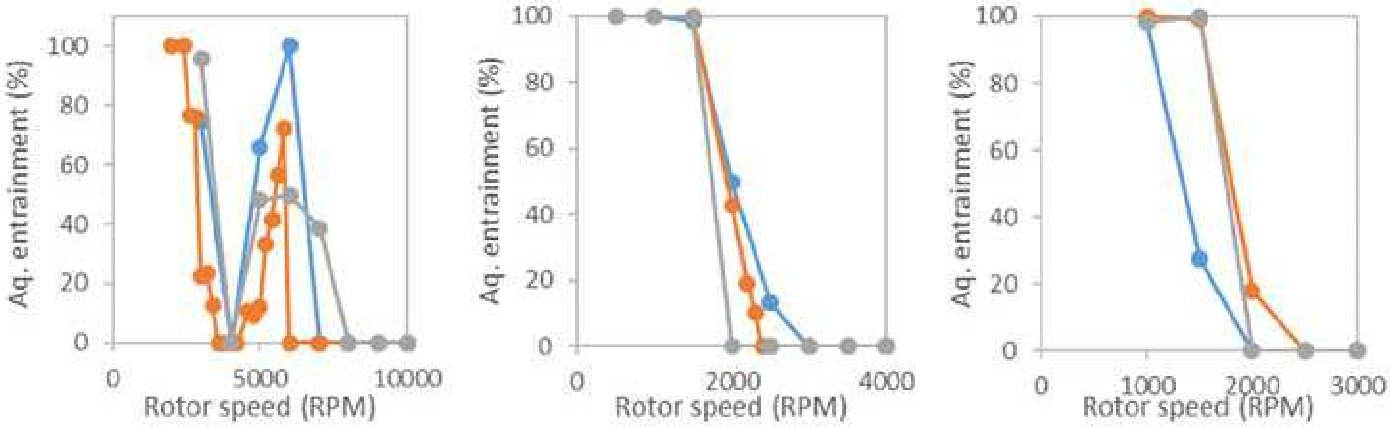


Figure 11: Rotor speed determination for inhibiting aqueous entrainment (%), left = 12mm RD ACC, middle -25mm RD ACC, right = 40mm RD ACC, grey = high S/A ratio, orange= equal S/A ratio, blue = low S/A ratio.

Figure 13: Cascade of 3 stages of 40 mm RD ACCs at the University of Leeds.

contactor, although the sediment layer sheared in the presence of aqueous throughput and resulted in the entrainment of solids through the cascade. The solvent extraction of nitric acid was also monitored and a decrease in extraction was observed [48].

In summary, AFCP has enabled the establishment of new ACC rigs at the University of Leeds, where experimental studies of hydrodynamics, solvent extraction, solids management, and on-line monitoring are in progress. AFCP is complementing experimental studies with computational fluid dynamics (CFD) modelling of flows inside ACCs.

2.1 Modelling work on ACCs

The hydrodynamic behaviour of centrifugal contactors is highly complex and not yet fully understood, in particular with respect to the intricate multiphase flow patterns and the droplet size distribution [47]. Computational fluid dynamics (CFD) has the potential to provide valuable insights on the multiphase flow observed within these devices [49]. However, the modelling of centrifugal contactors poses several challenges related to the complex nature of the flow field observed during their operation and to the presence of a broad range of interfacial scales; the latter, in particular, rules out the use of standard off-the-shelf multiphase flow modelling solutions to model ACCs [50, 51].

To address this issue, the University of Leeds has developed a GEneralized Multifluid Modelling Approach (GEMMA) which allows for the simulation of multiscale multiphase flows such as those observed in ACCs. A key feature of the approach is the ability to identify the local degree of resolution of the interfacial scales in each unit cell of the 'numerical mesh' of all the cells combined. An additional local switch variable C_α, representing a binary switch, is introduced:

- When $C_\alpha = 0$, GEMMA reduces to a standard multi-fluid model suitable for dispersed flow.
- When $C_\alpha = 1$, the interface is determinable; in those cells a novel formulation capable of guaranteeing a physically sound resolution of the interface deployed.

The modelling approach has been validated in standard multiphase flows where the GEMMA approach has been applied to the simulation of intensified liquid-liquid extraction including a lab-scale centrifugal contactor [52]. It has been shown that the modelling approach allows for an accurate prediction of the complex multiphase flow observed in centrifugal contactors, as qualitatively shown in Figure 14.

Notably, GEMMA makes available the key hydrodynamic parameters that control mass transfer within the device, such

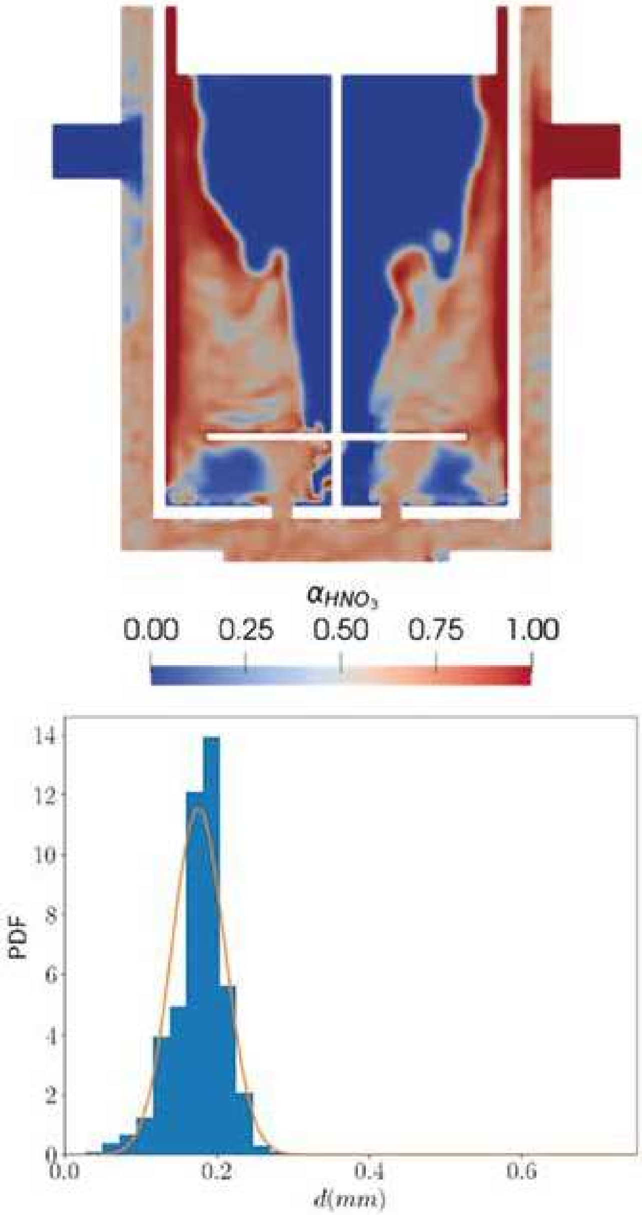


Figure 14: CFD simulation of a lab-scale centrifugal contactor: top – instantaneous aqueous phase volume fraction contours on the contactor mid-plane; bottom – droplet size distribution in the annular region.

as local hold-up, droplet size distribution, velocity fields, and residence time. This, in turn, has allowed for the development of an integral mass transfer modelling methodology which is informed by the results obtained by the detailed hydrodynamic CFD model. This approach has been validated in the simulation of acetone absorption [53] in a water/toluene/acetone system within a Rotating Disc Column (RDC). The hydrodynamic behaviour of the column, neglecting mass transfer, has been simulated in detail using the GEMMA approach. Successively, an integral mass transfer model has been built by representing the column as a series of Continuous Stirred Tank Reactors (CSTRs), with each CSTR representing one stage of the column. The mass transfer rate within each CSTR has been evaluated using the two-film theory [54] neglecting mass transfer resistance at the interface. The mass transfer coefficients in the aqueous and in the organic phases have been evaluated using the correlations of Treybal [55] and Laddha [56] respectively; the hydrodynamic parameters necessary for the evaluation of the mass transfer coefficients have been taken from the detailed CFD simulation. The results obtained for the solute concentration profiles in the aqueous and organic phases along the column height are in good agreement with the experimental measurements of Garthe [53] as shown on the left-hand side of Figure 15.

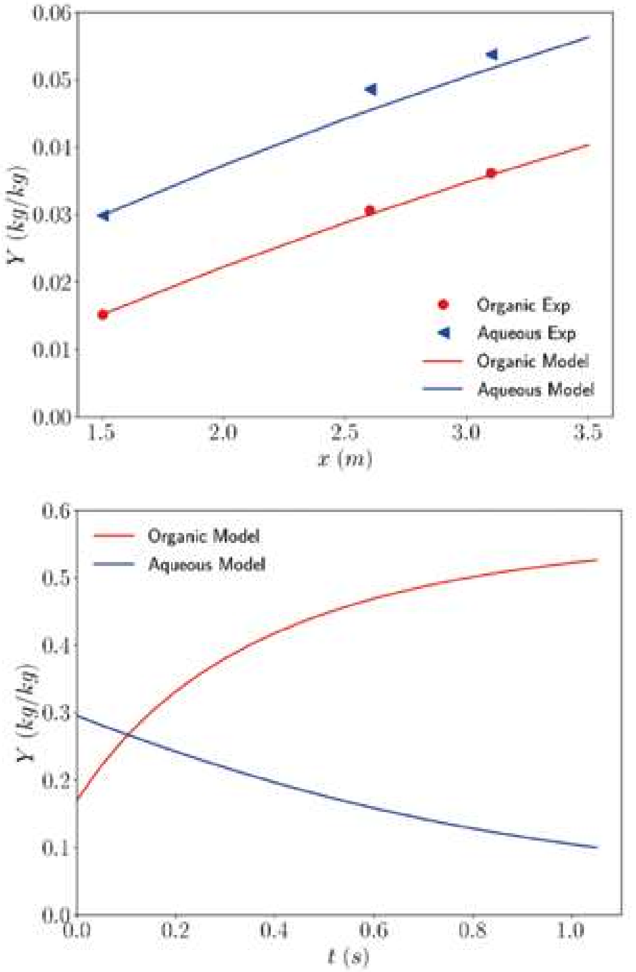


Figure 15: Top - Acetone mass fraction along the RDC Bottom - uranium mass fraction transient evolution in the ACC.

In addition, the same mass transfer modelling approach has been used to evaluate the mass transfer performance for uranium extraction in a HNO_3/TBP/$UO_2(NO_3)_2$ system in the same centrifugal contactor shown in Figure 14. The value of the distribution coefficient for the considered system, needed to evaluate the overall mass transfer coefficient in the two-film theory, is taken equal to 12 [57]. Only the annular region of the system has been considered in the mass transfer calculation, and this region has been represented using a single CSTR as a surrogate model. The change in the uranyl nitrate mass fraction in the two phases over time is obtained by integrating the CSTR governing equations over time and is shown on the right-hand side of Figure 15. The residence time in the mixing region of the ACC, evaluated from the CFD results, is equal to 1.1 seconds. The uranium content in the aqueous and organic phase inlets is assumed to be equal to 300 and 100 g/L, respectively, which corresponds to an initial $UO_2(NO_3)_2$ mass fraction ratio between the organic and the aqueous phases of 0.58. After 1.1 seconds, the mass fraction ratio has been observed to be equal to 5.3, compared to an equilibrium value of 12.

Future work will include the embedment of mass transfer modelling capabilities within the GEMMA approach, which would result in the capability of evaluating the mass transfer performance of the device locally, on a cell-by-cell basis, within the computational domain. Also, the simplifying assumptions taken in the evaluation of the mass transfer rate will be relaxed, in particular through the inclusion of finite-rate chemistry effects at the interface. Whilst this modelling approach is starting to provide insight into the fluid dynamics inside an ACC, the modelling approach is currently limited to two fluids; aqueous and solvent. Future expansion to three or four-phase systems; incorporating gas and solid phases. This expansion will allow the effect of rotor speed to be assessed, which is likely to be important when considering the effects of scale-up and change in rotor diameter caused by build-up of solids particle beds.

3. CONCLUSIONS

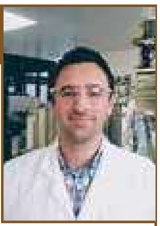
The technology gaps in our understanding of annular centrifugal contactors for the next generation of spent nuclear fuel recycling is being investigated in a modern setting. This article reports on the unique-in-the-UK capabilities that the University of Leeds's Nuclear Engineering Group has established, as a partner of the AFCP. Through the construction of a physical rig consisting of different sizes of ACC and computer modelling we are making progress towards the development of a 'digital twin'. The work is providing answers to the effects of scale-up from the lab to engineering scales.

One way to scale up to rotor diameters >200 mm likely to be required for any application in a future recycling plant, is to build a rig and test performance. Whilst this may well be necessary at some point, the complexity, time, and cost will be significant and so a quicker, less costly route would be of great benefit. An alternative is to build a digital twin using computational fluid dynamics (CFD), a technique used widely to predict the hydrodynamic behaviour of complex systems. A digital twin can be created that matches the 40 mm ACC in use at the University of Leeds and can be validated against it for the accuracy of performance prediction. Once validated, the model can then be applied to a CFD mesh of a full-scale ACC and the performance of plant-scale throughput can be predicted.

Our preliminary hydrodynamic studies have identified the flowrate/rotator speed conditions that allow efficient aqueous/solvent separation. Preliminary uranium extraction studies are still ongoing and will allow the effect of throughput upon stage efficiency. Future studies will allow the assessment of suspended particulates upon hydrodynamic and uranium extraction stage efficiencies.

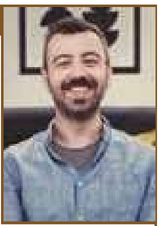
In summary, AFCP has enabled the establishment of a unique capability at the University of Leeds for testing and developing ACCs from laboratory to engineering scales and comparing experimental data to predictions from state-of-the-art CFD models. This capability can now play a vital role in proving the viability of ACCs for future recycling applications which would unlock substantial savings in plant size, costs, and waste generation as well as improvements in criticality safety and enabling the use of online analytical technologies for real-time nuclear materials accountancy and process control.

ALASTAIR BAKER, MCHEM PHD, MRSC FHEA



Alastair Baker gained an MChem Chemistry (2011) at Cardiff University/ Prifysgol Caerdydd where he also received his PhD (2016). Then undertook postdoctoral studies in catalyst development in self-optimising reactors between the Institute of Process Research and Development and the University of Nottingham. He has since commissioned the Leeds Nuclear Lab (2018) and undertaking process intensification studies from lab scale to pilot plant to deliver a viable next-generation aqueous recycling cycle for the UK.

ANDREA DE SANTIS, MSC, PHD



Andrea De Santis has a MSc in Nuclear Engineering from the University of Rome “Sapienza” (Italy) and a PhD in Computational Fluid Dynamics from the University of Sheffield (UK). He has held several positions in both industry and academia, where his work mainly focused on the simulation of thermal-hydraulics phenomena relevant to the nuclear industry. He is currently working as a data scientist in the private sector and a Visiting Researcher at the University of Leeds.

ALEX FELLS, MENG

Alex Fells is a chemical engineer from Norfolk, UK. He studied Chemical and Nuclear Engineering at the University of Leeds before working as a Process Design Engineer for Sellafield Ltd. He is currently pursuing his PhD with Prof. Bruce Hanson and Prof. Frans Muller at the University of Leeds, investigating the prediction of mass transfer in Pulsed Sieve-plate Extraction Columns.

TIMOTHY HUNTER, PHD

Timothy N. Hunter is an Associate Professor and head of the Nuclear Engineering Group (NEG) within the School of Chemical & Process Engineering, University of Leeds. He is a specialist in multiphase suspension, foam and colloidal systems, as well as process intensification. He has extensive experience in waste separation, slurry transportation and liquid effluent treatment.

BRUCE HANSON, MENG, PHD

Bruce C. Hanson holds a Leadership Chair in Nuclear Process Engineering at the University of Leeds and is an Honorary Professor of Nuclear Engineering at University College London. He is a named investigator on 10 grants, leading EPSRC’s ATLANTIC programme, and a Co-Director of the GREEN CDT. Before his appointment at Leeds, he spent 25+ years in the nuclear and chemical industries, across a range of functions, specialising in actinide separations and treatment of irradiated nuclear fuel. Most recently, he was the Technical Authority for Spent Fuel and Nuclear Materials at the National Nuclear Laboratory, where he a Visiting Senior Fellow.

CHRIS MAHER, PHD

Chris Maher was born in Cumbria, UK. He gained an MChem Chemistry at the University of York with radiochemistry at the University of Helsinki. He has worked at what is now the UK’s National Nuclear Laboratory since 2002 and has carried out work supporting the back-end of the nuclear fuel cycle. During this time, he studied part-time for a PhD at the University of Manchester (part of the EU FPVII ASCEPT project) relating to options for the head-end treatment of exotic nuclear fuels. Most of his time is spent working on the head-end treatment and dissolution of actinide-containing materials.

ROBIN TAYLOR, PHD

Robin is a Senior Fellow at the National Nuclear Laboratory (NNL) and Technical Lead for the Advanced Recycle and Sustainability theme of the Advanced Fuel Cycle Programme (AFCP). He is an Honorary Professor at the University of Lancaster and chair of the OECD-NEA Expert Group in Fuel Recycling Chemistry.

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ACRONYMS

AFCP	Advanced Fuel Cycle Programme
ACC	Annular Centrifugal Contactor
ACE	Annular Centrifugal Extractors
ACT	Advanced Centrifugal Contactor Test System
ANL	Argonne National Laboratory
ATF	Advanced Technology Fuel
BEIS	Business, Energy and Industrial Strategy
BNFL	British Nuclear Fuels Ltd
CC	Centrifugal Contactor
CFD	Computational Fluid Dynamics
CINC	Costner in Nevada Corporation
COTE	Commercial Off the Shelf
CPF	Coated Particle Fuel
CSTR	Continuous Stirred Tank Reactor
EIP	Energy Innovation Programme
GANEX	Grouped Actinide Extraction
GDF	Geological Disposal Facility
GEMMA	Generalized Multifluid Modelling Approach
G-force	Gravitational Force
HA	Highly Active
INET	Institute of Nuclear and New Energy Technology
JAEA	Japanese Atomic Energy Agency
MS	Mixer-Settlers
MSCR	MultiScale Contactor Rig
NNL	National Nuclear Laboratory
OK	Odourless Kerosene
PSEC	Pulse Sieve-Plate Extraction Column
PUREX	Plutonium Uranium Reduction Extraction
R&D	Research and Development
RD	Rotor Diameter
RDC	Rotating Disc Column
RR	Rousselet Robatel
S/A	Solvent to Aqueous Ratio
SNF	Spent Nuclear Fuel
SRS	Savannah River Site
TBP	Tri-Butyl Phosphate
THORP	Thermal Oxide Reprocessing Plant
UP2, UP3	Usine De Plutonium Plant No. 2,3