Spent fuel reprocessing and minor actinide partitioning safety related research at the UK National Nuclear Laboratory

Michael Carrott, Lauren Flint, Colin Gregson, Tamara Griffiths, Zara Hodgson, Chris Maher, Chris Mason, Fiona McLachlan, Robin Orr, Stacey Reilly, Chris Rhodes, Mark Sarsfield, Howard Sims, Daniel Shepherd, Robin Taylor, Kevin Webb, Sean Woodall, David Woodhead
National Nuclear Laboratory, Sellafield, Seascale, CA20 1PG, U.K.

Abstract

The development of advanced separation processes for spent nuclear fuel reprocessing and minor actinide recycling is an essential component of international R&D programmes aimed at closing the nuclear fuel cycle around the middle of this century. While both aqueous and pyrochemical processes are under consideration internationally, neither option will gain broad acceptance without significant advances in process safety, waste minimisation, environmental impact and proliferation resistance; at least when compared to current reprocessing technologies.

The UK National Nuclear Laboratory (NNL) is developing flowsheets for innovative aqueous separation processes. These include advanced PUREX options (i.e. processes using tributyl phosphate as the extractant for uranium, plutonium and possibly neptunium recovery) and GANEX (grouped actinide extraction) type processes that use diglycolamide based extractants to co-extract all transuranic actinides. At NNL, development of the flowsheets is closely linked to research on process safety, since this is essential for assessing prospects for future industrialisation and deployment. Within this context, NNL is part of European 7th Framework projects "ASGARD" and "SACSESS". Key topics under investigation include: hydrogen generation from aqueous and solvent phases; decomposition of aqueous phase ligands used in separations prior to product finishing and recycle of nitric acid; dissolution of carbide fuels including management of organics generated. Additionally, there is a strong focus on use of predictive process modelling to assess flowsheet sensitivities as well as engineering design and global hazard assessment of these new processes.

Introduction

Nuclear energy is seen as a key component in meeting future power demands as part of a balanced energy portfolio, both in the UK [1] and internationally. However, such demand requires an improvement in the efficiency of current fuel cycles to counter the expected increase in global demand for uranium and address proliferation and waste issues [2]. Future cycles will revolve around a new generation of fuel, requiring new technology for its manufacture and reprocessing, and will likely also involve closing of the fuel cycle to aid efficiency in the use of uranium alongside minimisation of the quantity of waste produced [3].

In Europe, focus is applied on advanced PUREX processes, including removal of Minor Actinides (MAs) and lanthanides from high level waste (HLW) followed by their separation (DIAMide EXtraction (DIAMEX) and Selective ActiNide Extraction (SANEX) processes), [4] and also the potentially more proliferation resistant Group ActiNide Extraction (GANEX) process [5]. Although the fundamental chemistry behind the separation concepts is still under research in many institutions, there is still much work needed to develop other areas related to the industrialisation of these closed fuel cycle approaches, including underpinning process safety. To this end, NNL is involved in several national and international research programmes.

The Safety of ACtinide SEparation proceSSes (SACSESS [6]) is a European Framework Programme 7 (FP7) programme that focuses on the safety and efficiency of separation processes and developing technologies for spent fuel treatment and management. NNL is working on projects that focus on hydrogen generation from aqueous and solvent phases in partitioning scenarios due to alpha-radiolysis of water; examining the decomposition of partitioning ligands in nitric acid to determine how effective their destruction is before recycling of the nitric acid; investigating flowsheet responses to maloperation scenarios; modelling of the separation process to determine speciation and distribution of actinides between phases and development of the safety assessment methodology to be implemented in these fuel cycles. The Advanced fuelS for Generation IV reActors: Reprocessing and Dissolution (ASGARD [7]) is a European FP7 programme that focuses on the research of the next generation of fuel to be used in future fuel cycles. NNL is working on projects that study the fabrication of these fuels and also the dissolution of the fuel for future reprocessing. Both these, and the previously mentioned SACSESS projects, are represented in the following sections, with the aims of the projects outlined and progress so far briefly highlighted.

Additionally, NNL is involved in supporting the ongoing reprocessing operations at the Sellafield site, including safety-related studies such as investigations of the detrimental effect of the residue of un-recycled ligands at the evaporator stage ("Red Oil"); responses to and causes of plant blockages; process optimisation and waste minimisation.

Contributions to the SACSESS project

Generation of hydrogen gas in separations processes

Generation of hydrogen gas is a considerable problem for the nuclear industry. Outside of long term storage issues, it can pose a problem due to the formation of flammable gas mixtures and pressurisation in containers. The radioactivity of the actinides and radioactive isotopes of other elements acts as a precursor for this hazard due to the radiolysis of aqueous and organic solutions and solids. As such, consideration of this situation is of concern for future partitioning scenarios, particularly where new ligands are proposed and higher burn-up fuels, such as fast reactor (FR) fuels, are to be reprocessed. The role of the nitrate ion is important due to its ability to scavenge radiolytic products before hydrogen gas is generated and its ubiquitous presence in process solutions. High quantities of nitrate ions can subsequently reduce the yield of hydrogen generation in comparison to that of pure water.

Although this process is documented, there has been little research performed on hydrogen gas generation by alpha-induced radiolysis at low concentrations of nitric acid. Such data would be of significant interest in safety assessments of reprocessing and minor actinide partitioning flowsheets [8-10]. Also of interest is the effect of the concentration of metal ions. Multivalent elements, such as plutonium, may affect the scavenging ability of nitrate ions.

NNL has performed a series of measurements on samples of plutonium in nitric acid, dosed with sulphate ions that were used to prevent the hydrolysis of plutonium at low concentrations of nitric acid. Reactions were performed in sealed vessels with an integrated syringe. Gas samples taken from these reactions were analysed by micro-gas chromatography, with resulting determined concentrations of hydrogen gas plotted as a function of time (Figure 1). From the results of varying concentrations of nitric acid, $G(H_2)$ values were calculated – the number of moles hydrogen produced per 100 eV of energy deposited into solution.

Figure 1: Generation of hydrogen gas from an 18.2 µmol solution of nitric acid containing plutonium nitrate
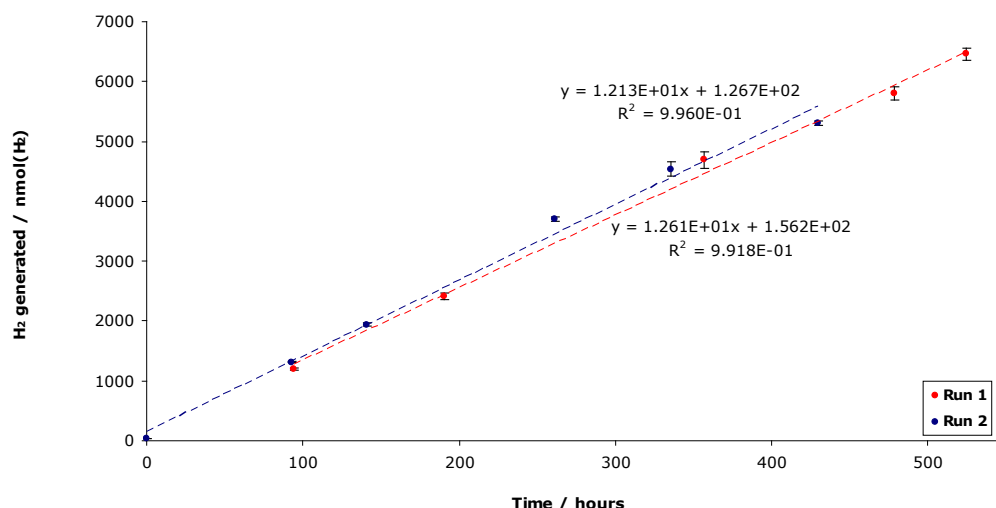
The experimental results depict a lower value of $G(H_2)$ than is observed in the literature, although trends are the same (higher hydrogen generation at lower nitric acid concentration). NNL is now studying the effect of concentration of plutonium to investigate this discrepancy, and to determine the effect of plutonium on the ability of nitrate to reduce hydrogen gas production. Future work will see experiments take place in organic solutions, specifically N,N,N',N'-tetraoctyl diglycolamide (TODGA) [11] solutions and also using americium as the alpha-source for the work.

Process modelling of the i-SANEX process

The innovative-SANEX (i-SANEX) process is one of the novel partitioning routes being assessed for future generation closed fuel cycles [12-14]. In this process, An(III) and Ln(III) are co-extracted from the fission products (FPs) in the PUREX raffinate. This is followed by separation of the actinide products from their lanthanide counterparts using selective stripping agents – a non-trivial step given the similar chemistry of the two groups of metals. Extractants that are being considered for use in these processes include DiGlycolAmides (DGAs), such as TODGA. One complicating factor is that the presence of high concentrations of metal can result in the formation of a third phase for solutions containing TODGA [12].

As such, an organic phase modifier is required to prevent this happening. TBP can be used to this end [12]. and is a strong candidate given its use in the PUREX process, although the i-SANEX process has recently been optimised with octanol as the phase modifier [15].

NNL is developing a process model of the i-SANEX process to enable the SACSESS community to simulate flowsheet tests and optimise flowsheet designs. The initial focus is on the extraction of nitric acid into the organic phase (TODGA + 5% octanol in a kerosene diluent). Previous work by Geist [16] revealed that [HNO$_3$] [2octanol] plays a significant role in attempting to define the speciation of nitric acid with octanol in the absence of other components. NNL has revisited the same dataset. Variation of the concentration of each component gives the equilibrium concentration by Equation (1).

$$\beta_{nm} = \frac{[nHNO_3.mOctanol]}{\gamma_{H^+}^n [H^+]^n \gamma_{NO_3^-}^n [NO_3^-]^n [Octanol]^m} \tag{1}$$

Plotting log([HNO$_3$]$_{(org)}$) against log[octanol] reveals the average stoichiometry to be [HNO$_3$].[1.4octanol], implying contribution of the 1:1 and 1:2 nitric acid:octanol products in the extraction of nitric acid into octanol in organic phases (along with high order octanol species *i.e.* [HNO$_3$].[1.4octanol]). The data fits well by comparison with experimental observations (Figure 2). Fitting leads to a 5% error by comparison with the experimental data, although it is higher when both acid and octanol concentrations are low.

Figure 2: Modelling of nitric acid extraction by octanol against experimentally observed data

394
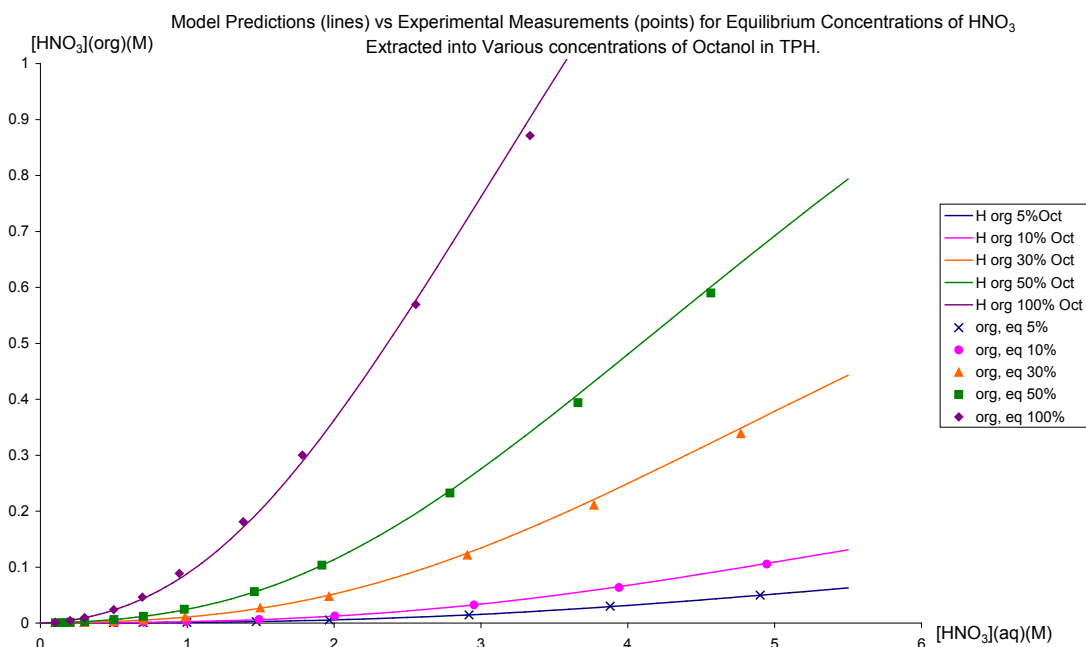
Work is in progress modelling the extraction of HNO_3 into TODGA in the absence of a phase modifier, with modelling based on datasets obtained from work at institutions including NNL and KIT-INE. The suggested speciation from modelling is $[HNO_3].[TODGA]$, but, at high acidities, higher ratios of HNO_3 are calculated (up to 3:1 ratio *vs*. TODGA). Preference was given to modelling data points at 0.2 M TODGA because of the relevance of this concentration for the i-SANEX process. Work will next focus on systems that contain nitrate complexes (such as those of americium and europium), and also modelling of nitric acid into the ternary system with octanol and TODGA.

Ligand decomposition experiments

After the purified actinides are stripped back into the aqueous phase in separations processes, the actinide nitrates are converted to oxides, typically *via* a precipitation stage. Nitric acid is then recycled to the head end of the process. However, questions arise as to the effects that the complexants used for stripping in the separations process may have on (i) the efficiency of the precipitation stage and (ii) process safety if recycled back into the head end of the process. Consequently, refluxing and evaporation may be required to concentrate actinide containing solutions and to destroy residual ligands. Previously, NNL has studied the decomposition of oxalate ions in nitric acid for the Thermal Oxide Reprocessing Plant (THORP) on the Sellafield site to ensure conditions are maintained for safe recycle of the concentrated oxalate mother liquor (CML) in the plant [17].

Similarly, it may be desirable to destroy the ligands used in the separation processes to prevent their interference with the conversion and recycling process. Under SACSESS, NNL is performing experiments on a variety of CHON ligands that have been proposed for future minor actinide separation schemes (Figure 3):

- Acetohydroxamic acid (AHA)
- Citric acid
- (2-Hydroxyethyl)-ethylenediaminetriacetic acid (HEDTA)
- Diethylenetriaminepentaacetic acid (DTPA)

Additionally, mellitic acid is being studied as this is a product of carbide fuel dissolution. The temperature and concentration of the nitric acid were varied to elucidate their effect on the decomposition of the aforementioned ligands by heating in a reflux experimental set-up for a set period of time. The samples were analysed by total carbon analysis – destruction of ligands should result in gaseous decomposition products, hence they should not be present in the sample after reaction.

Preliminary runs have taken place for all ligands of interest with the variables set at 60-119 °C for the temperature and 4-12 M for nitric acid concentration. Initial analysis of these results shows a less obvious relationship (in comparison to the studies of plutonium oxalate) [17] between the rate of ligand decomposition and the time exposed to destructive conditions (Figure 4). Further characterisation is to be performed on the samples to assess the fate of the ligands after reaction.

Figure 3: The ligands of study in the decomposition experiments

AHA

HEDTA

Citric acid

DTPA

Mellitic acid
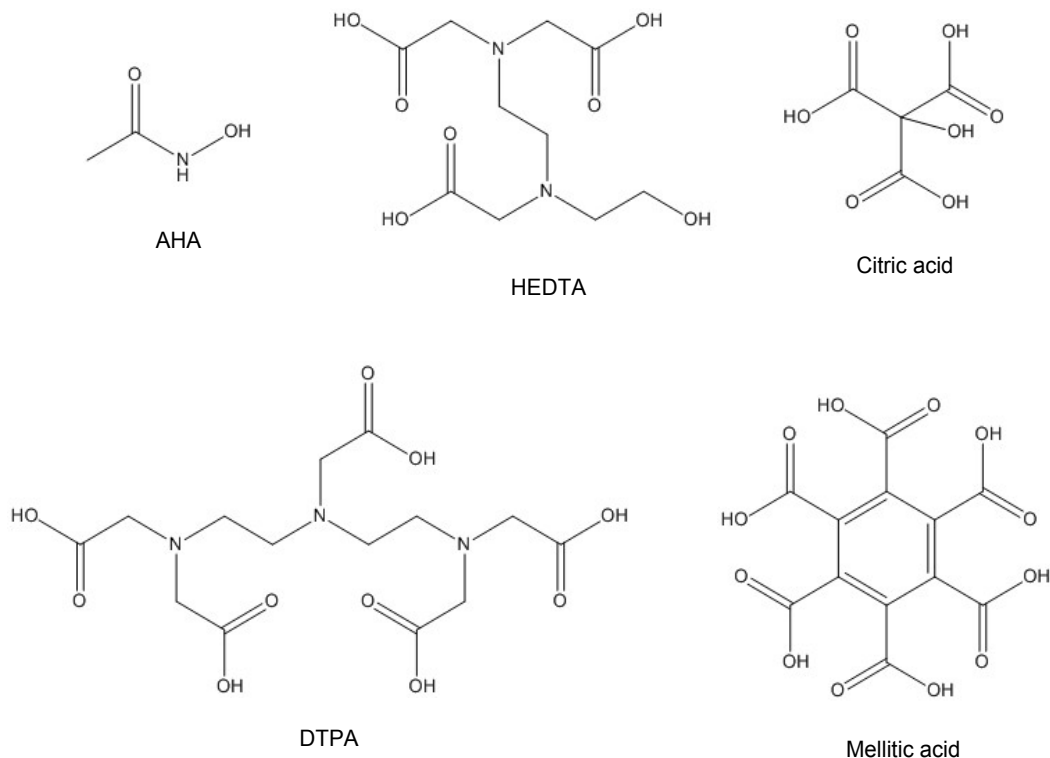
Figure 4: Total amount of carbon present in solution for partitioning ligands over 6 hours when reacted at boiling point in 12 M nitric acid
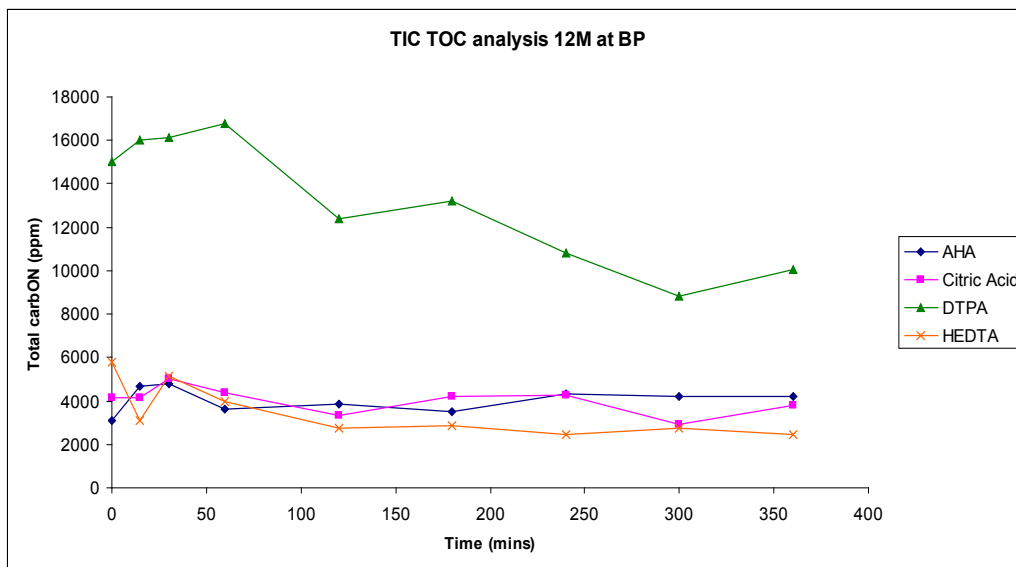

Production of SACSESS combined safety methodology

One of the goals of the SACSESS project is to produce a global safety review of an aqueous partitioning process. Before this can begin, the methodology for such a safety review needs to be

developed. This needs to be able to assess global safety of chemical separation flowsheets, allowing for future adaptations to industrial scale operations. It must also be able to meet International Atomic Energy Authority (IAEA) standards. The methodology seeks to determine the envelope of normal operations for a flowsheet when deployed at an industrial scale then determine the process hazards in both the normal operating regime and excursions from this envelope.

The methodology produced will follow international standards and lean on safety case experience from the nuclear industry and, as such, would be relevant to deployment of an assessed flowsheet in any country participating in the SACSESS program. NNL has performed a review of methodologies existing in these participating countries to start the methodology development. France and the UK are the only other European partners in SACSESS with operating reprocessing plants and up to date knowledge of the safety case basis of such plants. Therefore, this task is broadening the knowledge base across the SACSESS programme as to the type of analyses that must be performed to underpin process safety. The assessment work being undertaken will also inform the directions of the experimental and modelling safety studies to further support the goals of global safety work, evaluating and ensuring the basis for process safety.

The review identified two processes of considerable interest already established with the SACSESS community. The first of these is the Method of Organised and Systematic Risk Analysis (MOSAR) [18,19] approach (Figure 5). This is a technique used by the French nuclear industry which breaks down the system into a number of smaller target systems before systematically looking at a sequence of operations. It is a process evolved from the Analysis Method of Dysfunctional Systems (MADS) model and developed by the Commissariat à l' énergie atomique (CEA) in the 1980s.

Figure 5: Overview of the MOSAR methodology applied by the French nuclear industry
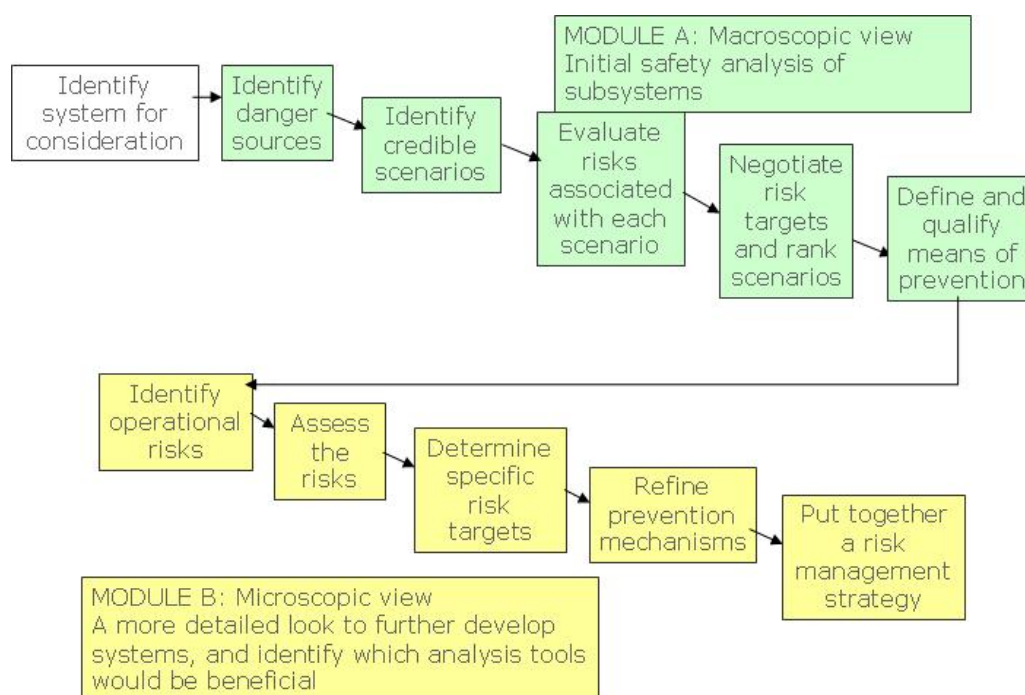
The second process of interest was the HAZard and Operability (HAZOP) study utilised by the UK chemical industry (and internationally, such as Japan).[20,21] This method provides a team based structural approach of analysing the design and operation of a plant and identifying potential hazards (chemical, radiological etc.) and breaks the process down into stages (known as HAZOP 0-2). The outcome of this review was that neither of these two approaches was directly applicable for SACSESS flowsheets because the processes are still at a relatively early stage of design. However, the node-based approach utilised by both is of use and has formed the basis of the methodology being developed for the global safety work.

The inaugral deployment of this methodology was undertaken for the EURO-GANEX process[22]. The chemical flowsheet for the chemical separation process was considered at a scale and throughput suitable for a demonstration or pilot facility (i.e. large enough that hazards would manifest in significant events). The process successfully explored the flowsheet and highlighted areas of vulnerability and the need to clarify the envelope of normal operations. The output will provide key recommendations for the R&D required to industrialise the flowsheet.

Further flowsheets are to be assessed using the methodology with the next flowsheet for consideration being the deployment of pyrochemical process.

Contributions to the ASGARD project

Uranium-Carbide fuel manufacture

Development of future closed nuclear fuel cycles will involve the use of different technologies than those in place currently, potentially including use of new fuel types. Two such fuels of interest are mixed uranium-plutonium-carbides and similar nitride fuels (MX fuels). (U,Pu)C fuels are under consideration for fast reactors. The fuel has a high fissile/fertile heavy metal content and a low fuel centre temperature in reactor, which means there is little restructuring in reactor and low release of fission gases and volatile fission products. (U,Pu)C fuel also has specific rod powers. These characteristics mean the fuel has high thermal conductivity yet low thermal expansion properties, which gives low fuel/clad mechanical interaction and reduced reactivity decline over the fuel lifetime. The high thermal conductivity may allow greater flexibility in fuel pin designs and improves plutonium breeding characteristics.[23] The main drawback of carbide fuels in-reactor is their relatively high irradiation swelling rate compared with oxide fuels, which can be an issue with regards to potentially damaging fuel-clad interaction. They are also more difficult to manufacture than oxide fuels as inert argon atmospheres are required for the process.

NNL is investigating the manufacture of carbide fuels, and is currently setting up custom-specification equipment in its Preston Laboratory to help development of a UC production flowsheet (Figure 6). (U,Pu)C fuel would be ~80% UC. Therefore, UC is a good lower radioactivity surrogate for performing manufacturing development work of (U,Pu)C. Cerium and/or neodymium can also be added as non-radioactive surrogates for plutonium and minor actinides, such as americium, respectively.

The new equipment being set-up for inert atmosphere operations includes milling, granulating, blending, pressing and a furnace for reduction and sintering. Work has now begun to calibrate the presses utilising previous NNL experience and using UO_2 as a precursor. In addition, a new particle size analyzer has been set-up and used to analyse potential raw material graphite powders for the production of UC. The candidate graphite powder identified as most suitable for initial manufacturing trials had a mean particle size of 19.5μm and a specific surface area of 5.9 m^2g^{-1}.

There has been a significant investment in microscopy and analysis equipment as part of the role of the Preston Laboratory in the UK Nuclear Fuel Centre of Excellence (NFCE) to allow analysis of grain and pore size distribution, stoichiometry and location and homogeneity of additives and surrogates. Future work will see investigations of these and utilisation of the inert atmosphere set-up to trial the UC flowsheet. Such work is scheduled to include density control investigations and also re-sinter testing to mimic in-reactor thermal behaviour.

Figure 6: The flowsheet deisgn for the production of UC fuel from UO₂ powder
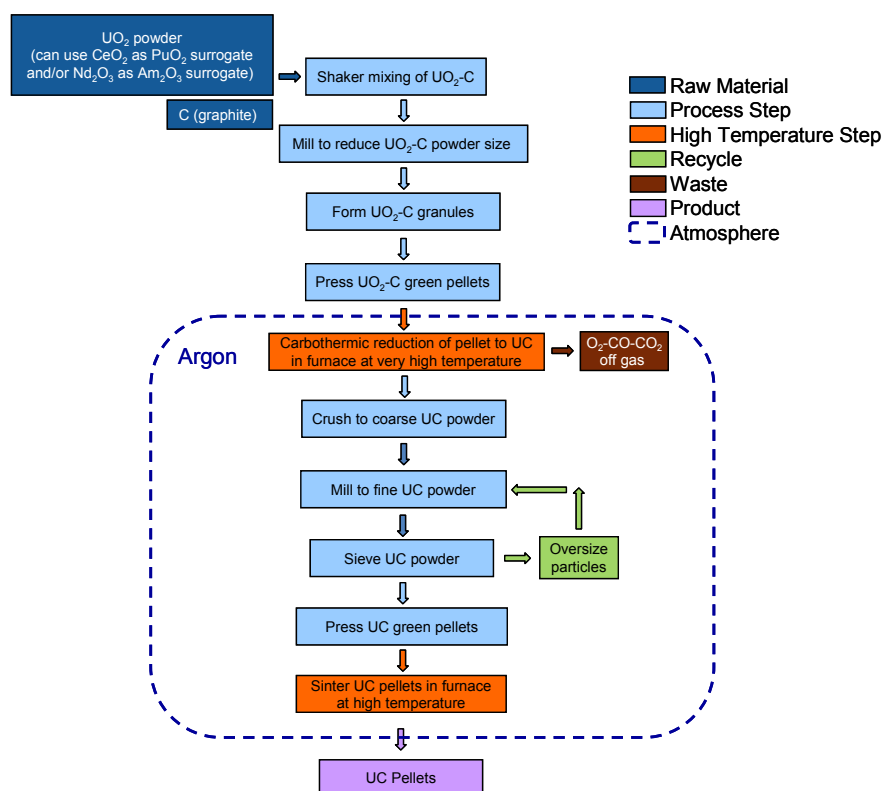
Uranium-Carbide dissolution experiments

One of the key objectives of a sustainable fuel cycle is that fissile material needs to be re-used and recycled. For next generation fuels, this may involve notably different processes than are already in place *e.g.* for UO₂ fuel. For (U,Pu)C fuel, there are currently two options:

- Direct dissolution of carbide fuel with further processing to give a solution suitable for separation processes.

- Pre-oxidation of fuel; conversion of the fuel to UO₂ to allow for dissolution and treatment.

NNL is focused on the direct dissolution of UC fuel under the ASGARD project as a surrogate for (U,Pu)C fuel, specifically aqueous dissolutions. Nitric acid is used allows for compatibility with current reprocessing strategies (*e.g.* the PUREX scheme). Reaction of UC in HNO_3 proceeds *via* Equation 2, leading to a mixture of solution, solid and gaseous products alongside unwanted organics in solution.

$$UC + xHNO_3 \rightarrow UO_2(NO_3)_2 + aNO_2 + bNO + cN_2O + dN_2 + CO_2 + \text{organics} \qquad (2)$$

Existing literature presents mixed reports on the quantity of carbon in solution[23]. NNL has performed work assessing this and other outputs, such as gaseous formation (none-CO_2 carbon-based gaseous products would have to be converted to CO_2 at a later date) by performing direct dissolution of unirradiated UC pellets (legacy fuel from the former UK fast reactor programme). The temperature and concentration of nitric acid were varied under inert atmosphere to allow measurement of off-gases.

Increasing reaction temperature resulted in an increase in reaction rate. Analysis of total carbon in solution indicates that there is also an increase in destruction of organics (see Figure 7). Increasing the concentration of nitric acid saw only a minimal effect on reaction rate, but a significant increase in destruction of organics. In addition, an as yet unidentified carbon based off-gas was produced. The increase in dissolved uranium was confirmed by an increase in the presence of uranyl(VI) in solution, confirmed by UV-vis spectroscopy.

Figure 7: The effect of temperature of nitric acid on dissolved carbon species (left) and the effect of temperature on the same (right)
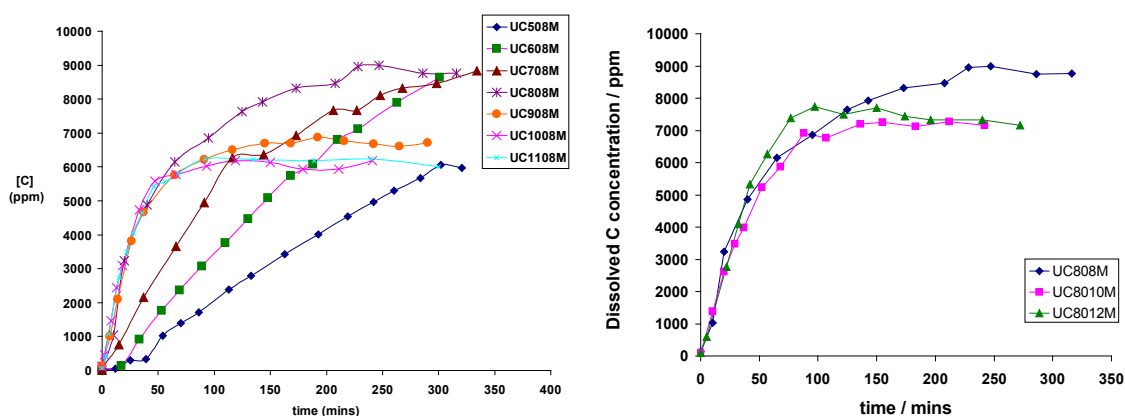
The legend for each graph depicts the temperature and concentration of each experimental run e.g. UC608M indicates a run at 60°C in 8 M HNO$_3$.

Analysis of the unidentified off-gas will continue. From these results the optimised conditions for UC dissolution conditions can be formulated.

Support to Sellafield; an exemplar safety-related project

"Red Oil"

"Red Oil" is the term given to highly exothermic reactions of reprocessing solvents with hot aqueous solutions (so called due to the colour of the residue at the end of the reactions). These reactions are violent, leading to uncontrolled gas and heat production. Such reactions have led to several incidents globally, such as:

- Savannah River, 1953 – Pilot uranium evaporator

- Ontario, 1980 – Uranium evaporator

- Tomsk, 1993 – hot solutions added to a storage tank.

In the last example, the result of the addition of 15 M hot nitric acid to organics in a storage tank was an explosion and a fire, damaging the building (Figure 8) and resulting in the spread of activity over a 130 km^2 area.

However, the actual mechanisms of "Red Oil" reactions are unknown. Such reactions are of considerable importance due to the role of TBP in current PUREX reprocessing and its potential use in future methods such as advanced PUREX and TODGA/TBP processes; hence the study of this reaction is of interest in underpinning safe operations of current and future actinide separations processes.

NNL is collaborating with London South Bank University (LSBU) on better characterising red oil chemistry. Experiments have taken place studying the effect of TBP in organic diluent in reaction with nitric acid. The reaction was monitored in a controlled scenario – solutions of nitric acid and TBP/OK were heated and resulting gas and heat production measured (Figure 9). The results of these studies have indicated that nitric acid also reacts with OK, but not as violently as high temperature reactions of TBP with nitric acid observed previously.

Future work will investigate the effect of metals on this reaction. This project is also interested in the role of ammonium nitrate in any reaction scheme – a compound which can end up in evaporators following reprocessing operations.

Figure 8: The aftermath of the explosion in Tomsk in 1993

Figure 9: Gas evolution data for the reaction of 10 M HNO$_3$ 4:1 20% TBP in OK at 80 °C.
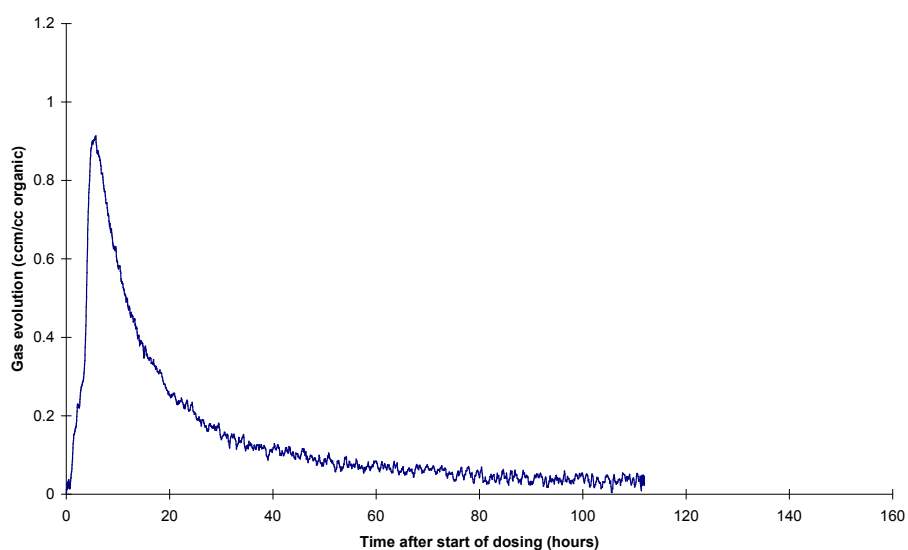
Summary

NNL is currently involved in European framework projects focused on development of options for future sustainable (closed) nuclear fuel cycles. Work on the ASGARD project has studied a potential future fuel in uranium-carbide, with work to commence on manufacture of the fuel. A comprehensive study has taken place of its dissolution in nitric acid to guide optimisation of reprocessing of the fuel.

Work on the SACSESS project has investigated many areas of separations process safety, from the detrimental effect of unwanted ligands and organics residing in solution, to the effect of radiolysis on the different phases and process modelling of actinide partitioning processes. NNL has also begun to address the issue of how to perform safety assessments on such processes that are still in the developmental stage.

NNL also continues to support safe and efficient reprocessing operations at Sellafield through, for instance, research on topics such as "red oil" reactions.

References

[1]   'Meeting the energy challenge, a white paper on nuclear energy', Dept. for Business, Enterprise & Regulatory Reform, January 2008, Crown Copyright, 2008.

[2]   'Nuclear fission', Energy Research Partnership Technology report, September 2010.

[3]   'Management of spent nuclear fuel and its waste', EASAC policy report #23, June 2014.

[4]   C. Ekberg, A. Fermvik, T. Retegan, G. Skarnemark, M.R.S. Foreman, M.J. Hudson, S. Englund, M. Nilsson, 'An overview and historical look back at the solvent extraction using nitrogen donor ligands to extract and separate An(III) and Ln(III)', Radiochim. Acta, 2008, 96, 4 (Europart 225).

[5   E. Aneheim, C. Ekberg, A. Fermvik, M.R.S. Foreman, 'Development of a novel GANEX process', Actinide and Fission Product Partitioning and Transmutation, 11th information exchange meeting, San Francisco, USA, 2010, 257-263.

[6]   http://www.sacsess.eu/

[7]   http://asgardproject.eu/

[8]   S. Bourg, C. Hill, C. Caravaca, C. Rhodes, C. Ekberg, R. Taylor, A. Geist G. Modolo, L. Cassayre, R. Malmbeck, M. Harrison, G. de Angelis, A. Espartero, S. Bouvet, N. Ouvrier, 'ACSEPT--Partitioning technologies and actinide science: Towards pilot facilities in Europe'. Nuclear Engineering and Design, 2011, 241, 3427-3435.

[9]   S. Bourg, 'SACSESS, a New Fp7 Euratom Collaborative Project on the Safety of Actinide Separation Processes', in ICAAP 2013 International Congress on Advances in Nuclear Power Plants. 2013: Jeju, Korea. Abstract FA076.

[10] B-G. Brodda, S. Dix, E.R. Merz, 'Radiolytic gas formation in high-level liquid waste solutions', Analyst, 1989, 114(3), 335-337.

[11] Y. Sasaki, Y. Sugo, S. Suzuki, S. Tachimori, "The novel extractants, diglycolamides, for the extraction of lanthanides and actinides in HNO3-n-dodecane system", Solv. Extr. Ion. Exc., 2001, 19, 91-103.

[12] M. Sypula, A. Wilden, C. Schreinemachers, G. Modolo, 'Separation of An(III) from PUREX raffinate as an innovative-SANEX process based on a mixture of TODGA/TBP', 1st ACSEPT international workshop, Lisbon, Portugal, 2010.

[13] G. Modolo, A. Wilden, P. Kaufholz, D. Bosbach, A. Geist, "Development and demonstration of innovative partitioning processes (i-SANEX and 1-cycle SANEX) for actinide partitioning, Progr. Nucl. Energ., 2014, 72, 107-114.

[14] G. Modolo, A. Wilden, A. Geist, D. Magnusson, R.A. Malmbeck, "A review of the demonstration of innovative solvent extraction processes for the recovery of trivalent minor actinides from PUREX raffinate, Radiochim. Acta, 2012, 100, 715-725.

[15] D. Magnusson, A. Geist, R. Malmbeck, G. Modolo, A. Wilden, 'A flow-sheet design for an innovative SANEX process using TODGA and SO3-Ph-BTP'. Procedia Chemistry, 2012, 7(0), 245-250.

[16] A. Geist, 'Extraction of nitric acid into alcohol:kerosene mixtures', Solv. Extr. Ion Exch., 2010, 28, 596-607.

[17] C. Mason, T.L. Brown, D. Buchanan, D. Morris, 'The destruction of oxalic acid in TPFL evaporators 1 and 2 under normal and potential abnormal conditions', NNL (08) 9481, Issue 4, 2008.

[18] Techniques de l'Ingénieur: MOSAR – Présentation de la Méthode, October 2003.

[19] Techniques de l'Ingénieur: MOSAR – Cas Industriel, April 2004.

[20] 'National Nuclear Laboratory Criticality & Safety Assessment (SEA) Section: Advisory Safety Memorandum SEA/SPR/GEN/347/10' Revision 1, NNL 10206 Issue 2, IMS-TECH-C&SA-GN-0005 Version 2: Information on Hazard and Operability Studies (HAZOPs), July 2010.

[21] Y. Kohato, S. Matsuoka, K. Tabeke, A. Takeuchi, K. Kuroiwa and K. Hayashi, 'Study on Application of Probabilistic Safety Assessment to Rokkasho Reprocessing Plant'.

[22] L. Flint, 'SACSESS deliverable D31.1a – a production of SACSESS combined safety assessment methodology, summary of NNL activity task 3.1.1', 2014.

[23] K. Bell, 'ASGARD milestone 4.2.1 – A literature review of carbide reprocessing', 2012.