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Process Description report of Concept Euro- GANEX Plant

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Concept Design of a Euro- GANEX Reprocessing Plant

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EXEC SUMMARY

As part of the GEN IV Integrated Oxide fuels Recycling Strategies (GENIORS) programme, the National Nuclear Laboratory (NNL) has been tasked with the production of a concept design of a Euro-GANEX reprocessing plant. This document outlines the design.

The design pack incorporates a flowsheet which provides mass balance data of species through the process. To enable this, a gPROMs model of the plant was constructed. FISPIN modelling data was used to provide a representative spent fuel inventory which acted as an input to the gPROMs model. The model output has been included as an appendix to this report whilst summaries have been included in Appendix 11.1.

The outcome of this report will provide a baseline for the Euro-GANEX process and will allow for safety evaluations and studies, after which further phases of design can commence. The baseline can also be utilised by the Sim-Plant tool, currently under development at NNL, which will allow for the comparison of the benefits of different reprocessing flowsheets.

GLOSSARY

AHA	Acetyl hydroxamic acid
AIROX	Atomics International Reduction Oxidation
ASTRID	Advanced Sodium Technological Reactor for Industrial Demonstration
BTP	Bis tryazinyl pyridine
CFA	Conditions For Acceptance
CML	Concentrate Mother Liquor
CVF	Constant Volume Feeder
DEHIBA	N,N-di-2-ethylhexyl-isobutyramide
FISPIN	Spent fuel inventory modelling software
FP	Fission Product
FR	Fast Reactor
GANEX	Group ActiNide Extraction
GDF	Geological Disposal Facility
GEN IV	Generation 4 Nuclear Reactors
GFR	Gas-Cooled Fast Reactor
HA	High Active
HLW	High Level Waste
HM	Heavy Metal
ILW	Intermediate Level Waste
INL	Idaho National Laboratory
KAERI	Korean Atomic Energy Research Institute
LA	Low Active
LFR	Lead-Cooled Fast Reactor
LLW	Low Level Waste
MA	Mixed Actinide
MEB	Multi Element Bottle
MEO	Mediated Electrochemical Oxidation
MOX	Mixed Oxide
MSM	Master Slave Manipulator
NNL	National Nuclear Laboratory
NOX	Nitrogen Oxides
OK	Odourless Kerosene
OML	Oxalate Mother Liquor
OREOX	Oxidation Reduction Oxidation
PCM	Plutonium Contaminated Material
RFD	Reverse Flow Diverter
ROE	Red Oil Excursion
SNF	Spent Nuclear Fuel
SS	Stainless Steel
TDN	Thermal DeNitration
THORP	Thermal Oxide Reprocessing Plant
TR	Thermal Reactor
UN	Uranyl Nitrate
Volox	Voloxidation

CONTENTS

Exec Summary	2
Glossary	3
Contents	4
1. Introduction	7
2. Functional Specification and Design Basis	8
2.1. Flowsheet	8
2.2. Battery Limits	8
2.3. Basis of Operation and Maintenance	9
2.4. Effluent Strategy	9
2.5. Location	10
2.6. Operational Shielding	10
2.7. Criticality Control	10
3. Fast Reactor Fuel – Considerations for Concept Design	12
3.1. Construction	12
3.2. Pu Content	14
3.3. Post-Irradiation Cooling and Storage	14
4. Process Description	16
4.1. Head End	16
4.1.1. Shearing	16
4.1.2. Voloxidation	17
4.1.3. Dissolution	17
4.2. Chemical Separations	19
4.2.1. EURO GANEX	19
4.3. Uranium Finishing	24
4.4. Mixed Actinide Finishing	25
4.5. Key Equipment List	26

5. Technical Optioneering	27
5.1. Head-End Operations	27
5.1.1. Pre-Treatment of Fuel	27
5.1.2. Fuel Dissolution	30
5.2. Chemical Separations	38
5.2.1. Solvent Extraction	38
5.3. Finishing	42
5.3.1. Uranium Finishing	42
5.3.2. Mixed Actinide Finishing	42
6. Waste and Effluent	45
6.1. Solid Waste	45
6.2. Liquid Effluent	45
6.3. Gaseous Effluent	45
7. Flowsheeting	46
7.1. Fuel Feed	47
7.2. Model Flowsheet	49
8. Conclusions	50
9. Recommendations	51
9.1. General Recommendations	51
9.2. Flowsheeting Recommendations	52
10. References	53
11. Appendices	55
11.1. Summary Tables	55
11.2. Flowsheeting Model Assumptions Register	66
11.2.1. General Assumptions	66
11.2.2. Acid Dissolver	66
11.2.3. Acid Wash	66

11.2.4.	Density Model (Aq, Gas, Or)	67
11.2.5.	Buffer Tank.....	67
11.2.6.	Calcine.....	68
11.2.7.	Centrifuge	68
11.2.8.	Evap (U Finishing).....	68
11.2.9.	Evap_Ox (MA Finishing)	69
11.2.10.	Fuel_Feed.....	69
11.2.11.	Fuel Prep (Voloxidation)	69
11.2.12.	Mixer (CDTA).....	70
11.2.13.	NO _x Conditioning	70
11.2.14.	Ox_Filtration (MA Finishing)	70
11.2.15.	Precipitation (MA Finishing)	71
11.2.16.	First_1_Cycle (Primary Separation).....	71
11.2.17.	First_2_Cycle (U Backwash)	71
11.2.18.	Second_1_Cycle (TRU Scrub)	72
11.2.19.	Second_2_Cycle (Actinide Strip)	72
11.2.20.	Second_3_Cycle (Lanthanide Strip)	73
11.2.21.	Shearing	73
11.2.22.	Solvent Addition (First and Second cycle).....	74
11.2.23.	Solvent Recycle (First and Second cycle)	74
11.2.24.	TDN_System.....	74
11.2.25.	Valency_Conditioning	75

1. INTRODUCTION

The European Horizon 2020 GENIORS project combines the efforts of 24 European partners to improve: the current recycling of spent nuclear fuel, and future recycling strategies to be implemented in the 4th generation of nuclear reactors (expected to enter operation by 2030) [1].

Euro-GANEX (Grouped Actinide Extraction) is an advanced aqueous reprocessing flowsheet developed as part of the European Union's funded SACSESS project [2]. Though it is broadly similar to the widely used PUREX process, the GANEX process utilises different extraction chemicals to extract a transuranic actinide product rather than a pure plutonium product. The benefits of this type of extraction are two-fold: the absence of pure plutonium in the product increases its proliferation resistance and the smaller quantities of actinides sent to high level waste (HLW) significantly reduces the heat loading of waste packages. Full recycling of actinides can result in a reduction in the time required for high level waste (HLW) to reduce to reference levels from ~100,000 years to ~500 years [3]. This increases the quantity of waste that can be incorporated per HLW container, reducing overall strains on storage facilities.

This document outlines the concept design of an advanced reprocessing plant, operating a Euro-GANEX flowsheet, to reprocess spent Gen IV fast reactor mixed oxide fuel. This design builds upon previous concept design work packages [2] [4] performed by NNL for the GANEX process and has incorporated learning and key information where appropriate. A flowsheet has also been produced to compliment and inform this design (see Appendix 11.1). This provides pertinent data, such as mass and volume flowrates.

Throughout the conceptual design, key issues and potential options for future stages of design (preliminary, detailed etc.) are identified. The exact technology utilised in the plant design is expected to change as the design develops and technology within the sector matures.

This design will be used as the basis for a safety assessment that will be conducted as part of Work Package 9 (Process Safety).

2. FUNCTIONAL SPECIFICATION AND DESIGN BASIS

2.1. FLOWSHEET

The Euro-GANEX variant of the GANEX process has been selected as the basis for this concept design. This is due to the fact that the Euro-GANEX flowsheet has been selected as a baseline flowsheet for the GENIORS programme.

The design will be for an industrial scale hydrometallurgical (aqueous) reprocessing plant with a fuel throughput of five tonnes heavy metal (pre-irradiation) per day (5 tHM/d or 1000 tHM/yr). This is analogous to the scale of the THORP and La Hague reprocessing plants and is therefore anticipated to be close to the throughput of a next-generation reprocessing plant.

The fuel to be reprocessed shall be of the FR MOX variety. Where possible, this design shall utilise the concept ASTRID fuel as a reference. Where it is not possible to draw information from the ASTRID fuel, experience from similar fuels e.g. the Dounreay fast reactor programmes will be used. This basis was agreed by consortium members at the first six-monthly GENIORS meeting in Prague in November 2017.

The exact fuel composition used for flowsheeting purposes can be found in section 7.1.

Figure 1 outlines the scope of the GENIORS concept EURO-GANEX reprocessing plant and general fuel recycling strategy. Boxes within the blue area are to be considered within the concept plant, in-keeping with the GENIORS battery limits (from pond to powder product).

Figure 2 illustrates the generic block flow diagram for the concept plant, identifying each of the major unit operations. This provides a basis for discussions in the technical optioneering and process description sections, where (respectively) the potential technologies to be used and a detailed description of the concept plant are presented.

2.2. BATTERY LIMITS

The design will include the operations that would normally occur within the reprocessing facility and exclude ancillary plants such as waste treatment facilities.

A fuel to fuel approach will be taken, whereby the design looks at most operations from spent cooled nuclear fuel to the production of powder feed for fresh fuel production. The design will include head-end operations such as fuel shearing, dissolution in nitric acid, and feed clarification. Chemical separation, including solvent washing and recycle, as well as fuel finishing processes. It is assumed that there will be fuel cooling and storage facilities adjoined to the reprocessing plant, however this will not be considered within the scope of this design. The facility to receive, handle and dismantle spent fuel assemblies (as appropriate) is included.

The feed to the process will be spent Gen IV fast reactor (FR), Stainless Steel (SS) clad, Mixed Oxide (MOX) fuel. As little data exists concerning the spent fuel composition of a commercial FR MOX fuel pin a mock-fuel pin was constructed using FISPIN model data. The mock-fuel utilised a mixture of Driver and Breeder fuel to create a fuel representative of what would be reprocessed in reality. See section 7.1 for more details on the flowsheet feed.

The fuel feed processed in the head-end section of the plant will exit as dissolver product solution and feed into the first cycle of the chemical separation section. The actinide product solution from the first cycle will then form the feed for the second cycle where actinides are separated from the fission products and lanthanides, which can act as neutron poisons, to produce suitable product for fuel manufacture.

Two major products are obtained through the process, these are:

- UO_3 powder product (from the first cycle)
- A Mixed Actinide (MA) powder product (from the second cycle)

All products are assumed to meet the conditions for acceptance (CFA) for the stores on the host site.

The wastes of the system will be:

- A Lanthanide loaded aqueous stream
- A fission product (FP) loaded aqueous stream
- Two degraded solvent waste streams (from solvent purges)

The treatment and disposal of these wastes will not be considered as part of this design, however significant safety considerations regarding their handling should be noted.

2.3. BASIS OF OPERATION AND MAINTENANCE

The shearing of fuel into the head-end process will be performed in a semi-batch wise manner and will take the form of several campaigns per year. Fuel assemblies will be fed one after the other to satisfy the throughput requirement of the downstream dissolvers and the availability of feedstock fuel. The plant will operate for 200 operational days per year and a 24 hour mode of operation is assumed during campaigns.

During campaigns the process downstream of shearing operations will be operated in a continuous manner, including fuel pre-treatment and the dissolvers. Prior to chemical separation, two buffer tanks will hold a days' worth of feed liquor to ensure a normalised supply of feed to the chemical separation part of the process. The buffer tanks will operate in an alternate fashion; as one is full and undergoing sampling (to ensure the feed meets plant requirements) the other will be filled. This technique of employing two buffer tanks will also be used between each unit on all of the waste streams. This enables the units to be decoupled from one another, enabling sufficient time for sampling.

The plant will comprise a fully welded stainless steel piped system and all pipe and fittings will conform to the applicable nuclear industry piping specification. The grade of steels used should be matched to the structural and corrosive requirements of each unit.

A "remove and replace" maintenance philosophy has been adopted for this design due to the complexity of process equipment within the cells, and dose uptake of workers associated with in-cell operations.

2.4. EFFLUENT STRATEGY

Solvents will be continuously cleaned and recycled within the solvent wash and recycle section of each GANEX solvent extraction cycle. All effluents, including purged solvent from the solvent washes, will be discharged to

the appropriate waste treatment facility located on the same site. Effluents will be sampled and monitored to assure they comply with the waste plant's CFA.

2.5. LOCATION

It is assumed that the process outlined in this design will be contained within a single facility which may have additional ancillary plants, not considered in this design, adjoined. This facility will be located on a licenced nuclear site. It is also assumed that this site will host the entire supporting infrastructure and supporting plants which complement the reprocessing facility e.g. waste management complexes and storage, continuous power, process steam, fuel storage, a transport network, etc.

2.6. OPERATIONAL SHIELDING

The plant will be divided into cells according to unit operation, each with the appropriate level of radiation shielding. At this point in the design, in the absence of a shielding and dose assessment, the shielding on each cell/area of plant must be pessimistic i.e. each cell is heavily shielded. This can be refined at later phases of design to balance cost and risk.

Remote operations, such as powered Master Slave Manipulators (MSMs), will be utilised where possible to provide an operator interface whilst also minimising operator dose uptake. This technology has been proven extensively within the nuclear industry.

2.7. CRITICALITY CONTROL

Due to the high throughput of fissile material, a criticality philosophy must be considered for this design. A safe-by-shape philosophy will be employed where possible, which dictates the maximum geometry of process equipment and storage tanks. Where this is not possible a safe-by-mass strategy, whereby the quantity of material permitted to flow/accumulate at any one time cannot possibly initiate a criticality event, will be adopted. For now this will be taken as a bounding case. A full criticality overview of the Euro-GANEX process will be provided by IRSN in Task 93 of WP9 of GENIORS.

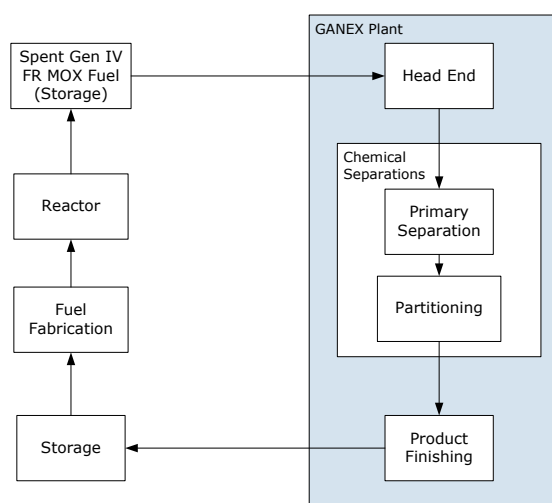


Figure 1 - High Level BFD

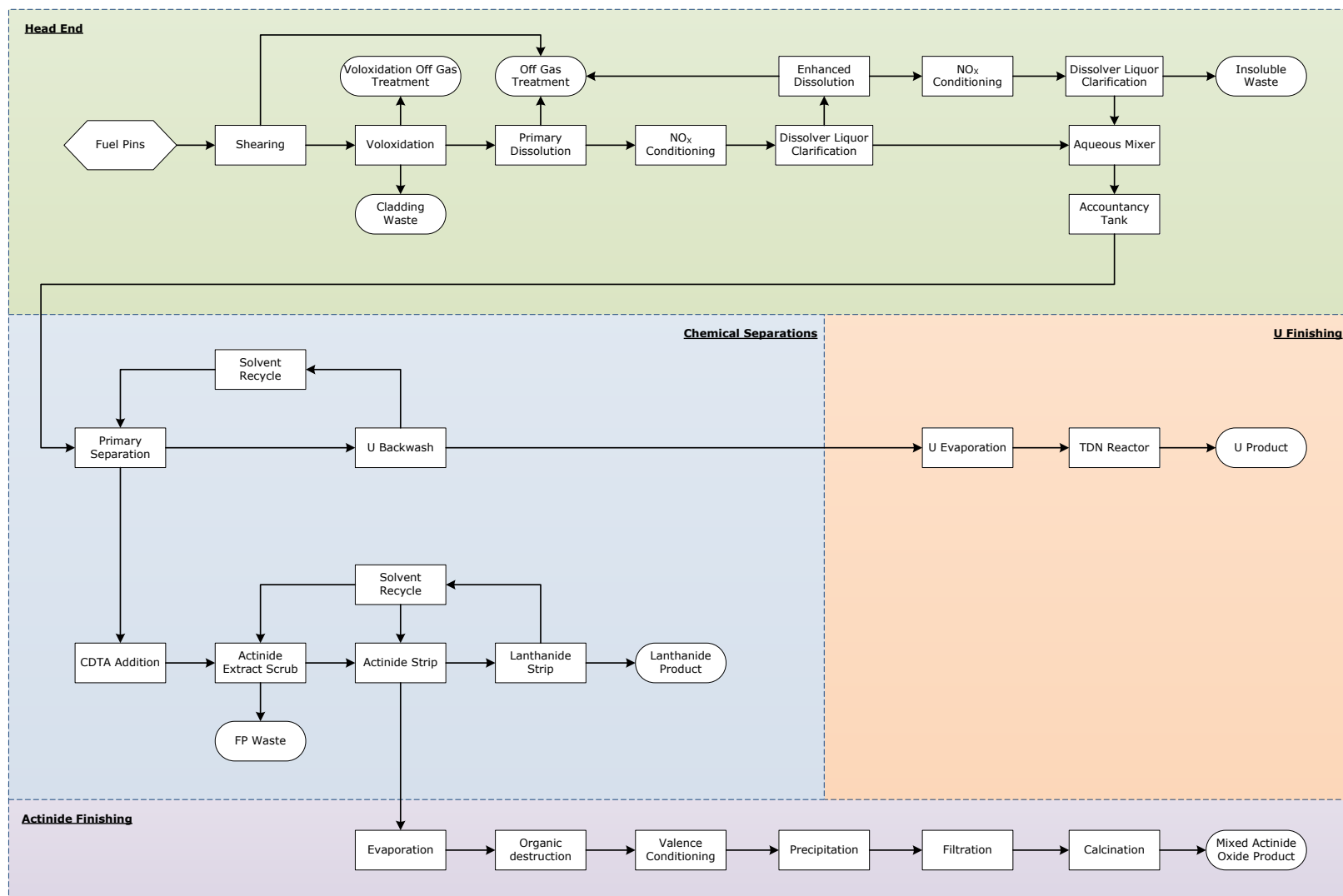


Figure 2 - Overall Euro-GANEX Process BFD

3. FAST REACTOR FUEL – CONSIDERATIONS FOR CONCEPT DESIGN

3.1. CONSTRUCTION

The key difference between TR MOX and FR MOX from a reprocessing perspective is the design of the fuel assembly and how this influences the head end design of the plant.

The Advanced Sodium Technological Reactor for Industrial Demonstration (ASTRID) is a concept under development by the Commissariat à l'énergie atomique et aux énergies alternatives (CEA). It is envisaged as a 600MWe prototype of a 1500MWe commercial scale Sodium-Cooled Fast Reactor (SFR) to be deployed by 2050. This concept design will take the design of a commercial ASTRID fuel assembly as a reference fuel [5]. It should be noted that the fuel design is only indicative of what a commercial fuel assembly may look like.

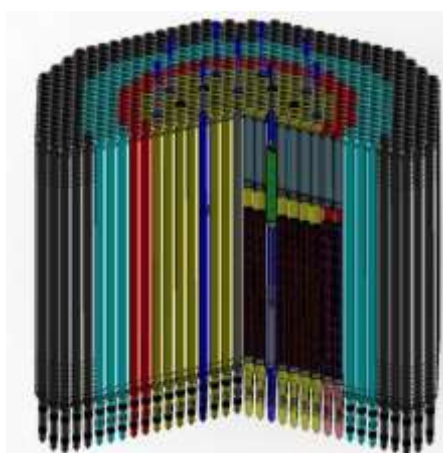


Figure 3 - Concept ASTRID reactor core [6]

The concept fuel assembly will comprise of 331 stainless steel fuel pins, each containing from 800-1000mm of fuel (in length) depending upon the position of the pin in the reactor core (Figure 4 below illustrates a 217 pin assembly). These are clad in a hexagonal stainless steel sheath incorporating flow channels for the sodium coolant. The fuel pins are 9.7mm in diameter, which can be narrower than TR fuel pins (which vary greatly typical diameters are 9.5mm - PWR and 14.5mm - AGR) [7] [8], and will be spaced apart using metal wire. FR fuel is much heavier than TR fuel with a cladding to fuel ratio of approximately 2.7. The average fuel assembly will comprise of ~190kg HM and ~517kg cladding. The sections of the fuel assembly above and below the fuel pin bundle house various nozzles to allow for a flow of sodium. Lifting lugs and other mechanisms to lock the assembly in place in the reactor are also present. Figure 4 and Figure 6 illustrate the concept ASTRID fuel.

It would not be appropriate to assume that, like a TR fuel assembly, the fuel can be chopped as a single entity by a guillotine blade. This operation would be very intense for the equipment involved, potentially creating excessive wear and damage. As a result the fuel pin bundles should be separated from the outer cladding prior to mechanical shearing, potentially via some form of cutting. This activity would also reduce the quantity of metal clad unnecessarily put through the dissolver. The remaining outer cladding may need to undergo size reduction operations prior to disposal via encapsulation.

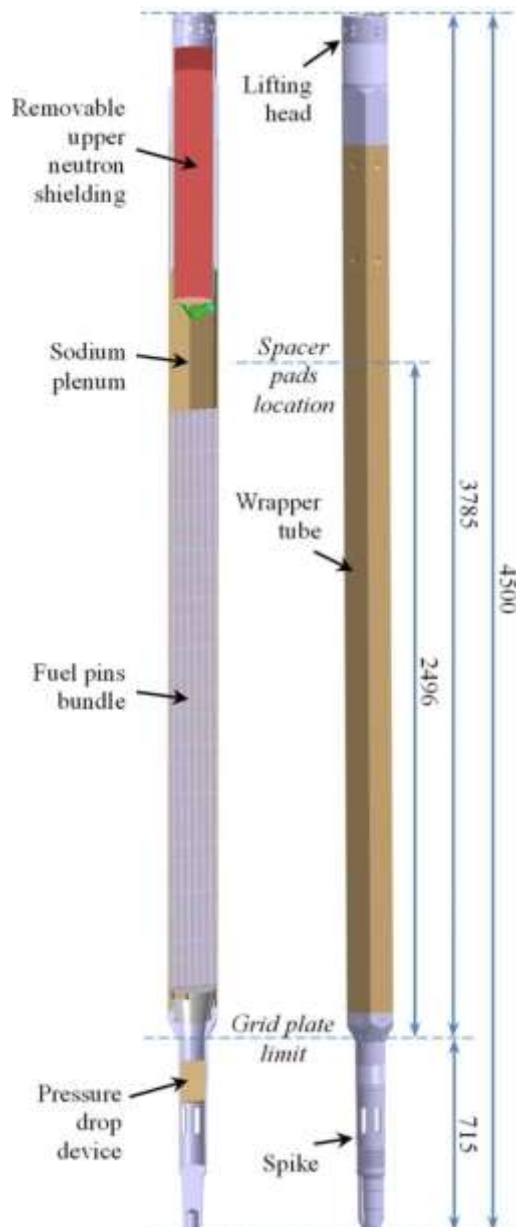


Figure 4 - Cross section of an ASTRID CRF v4 fuel assembly (dimensions in mm) [5]

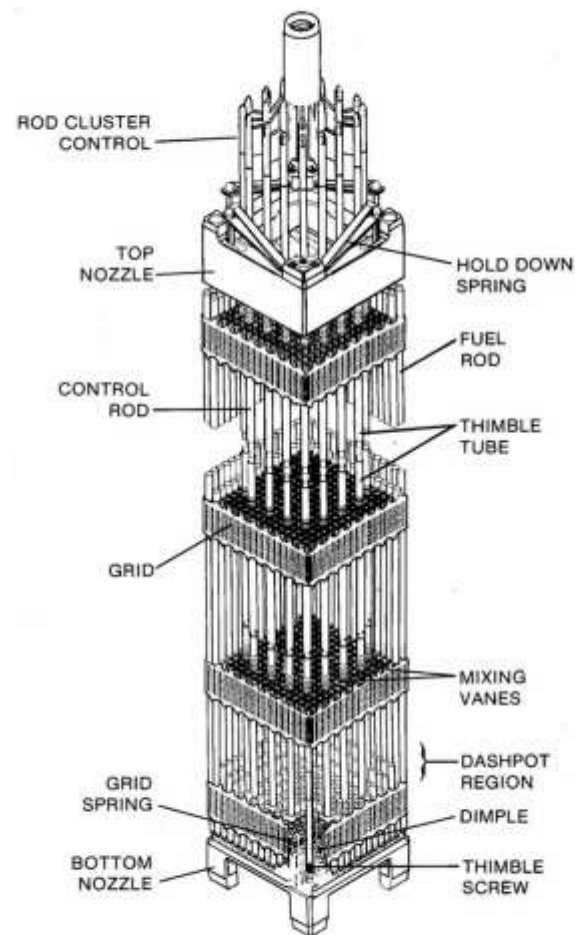


Figure 5 - PWR 17x17 pin assembly (for comparison) [9]

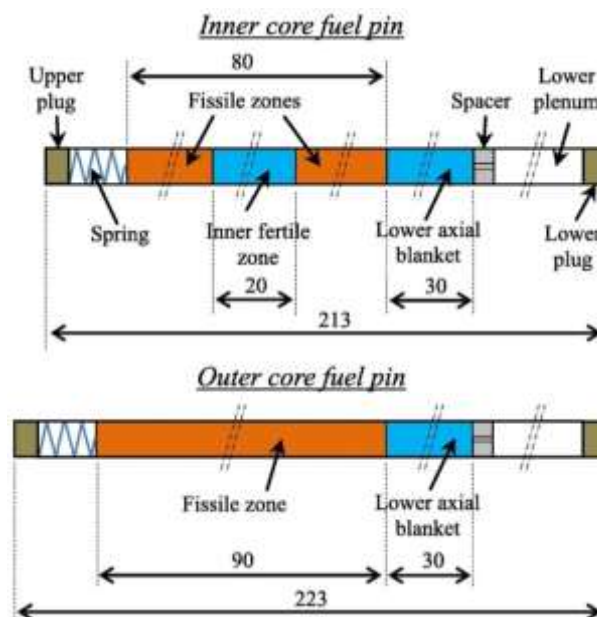


Figure 6 - Cross section of ASTRID fuel pins (dimensions in cm)

3.2. PU CONTENT

FR SNF will ultimately contain much higher concentrations of plutonium (approximately an order of magnitude more than TR SNF) [10], resulting from higher initial Pu concentrations, higher burnups, and shorter cooling periods (less than two years expected) prior to reprocessing. Therefore FR SNF is associated with much higher radioactivity.

The higher Pu content of FR fuel has an impact on the criticality management of the plant, especially in the head end dissolution stages where the absence of protective measures could result in a serious criticality event. The higher Pu concentration also has a detrimental effect on dissolution rates, resulting in a higher concentration of Pu-rich solids. Care must be taken when designing these sections of plant so that a critical mass cannot accumulate, whether this is by the control of mass entering the system, or restricting the volume that the material can occupy.

Spent FR fuel is much hotter (both in a radioactive and thermal sense) than spent TR fuel. The design will have to accommodate this through the use of additional shielding and remote handling equipment. Radiometric monitors will also have to compensate for the higher radiation levels expected, modelling simulations will likely play a key role in ensuring that the safety systems of the plant can effectively detect safety events in good time.

3.3. POST-IRRADIATION COOLING AND STORAGE

The type of SNF storage adopted will affect the type of storage facility adjoining the reprocessing plant and consequently the handling and unpacking stages which prepare the fuel for reprocessing. There are three potential ways in which spent SFR fuel can be stored [11]:

- **Storage in sodium** – The fuel is placed into a storage container containing a pool of molten sodium and the temperature is maintained between 150-200°C. An internal fuel storage container could be utilised if required. Fuel will undergo a washing stage prior to reprocessing to remove any residual sodium which would react with the aqueous liquors in the dissolver.
- **Dry storage** – In this scenario the fuel would be placed in an internal container within an inert gas flow (either forced or passive) for cooling. A pre wash stage would be required at the reprocessing plant to remove residual sodium from the fuel.
- **Wet storage** – This strategy involves the removal of residual sodium prior to storage at the reactor site. A steam-nitrogen process may be suitable though further investigation would be needed at a later design stage. The fuel is then placed into a container suitable for water-cooled storage in ponds.

Each of these options has an impact on the head end design of the reprocessing plant. To enable the progression of this design it will be assumed that the fuel will be wet stored having been thoroughly cleaned of all sodium residue at the reactor site. The dangers of introducing sodium into the aqueous reprocessing facility should however be emphasised.

4. PROCESS DESCRIPTION

This section outlines the proposed concept process, taking into account the technology options discussed in section 5. All four major sections of the plant are covered, with overviews of each unit operation. Block flow diagrams of the process can be found in section 11.1.

4.1. HEAD END

4.1.1. SHEARING

Fuel will arrive at the reprocessing site and be stored within Multi Element Bottles (MEBs) in the cooling pond facility adjoining the reprocessing plant. It is expected that the pond(s) will have a nominal total storage capacity of 3000 tU [12]. The facility shall be constructed in a modular fashion allowing for expansion if required. Fuel will be removed from the MEB, transferred underwater using a remote fuel handling machine and transferred to the shearing equipment cave via a sloped elevator.

A facility to prepare fuel assemblies prior to shearing is required. This will utilise remote handling devices such as MSMs to remove the outer cladding of the fuel and extract the fuel pin bundle for shearing. Further investigation is required to determine the effects of shearing a fuel bundle as a single array on the crimping of pins and dissolution rates (see section 5.1.1 for more information). It is assumed that additional dismantling of the bundle will be required so the preparation cell will include this capability. The outer cladding will require cutting, to be conducted using a cutting disc. Figure 7 illustrates the cutting of a FR assembly by CEA and AREVA [13].

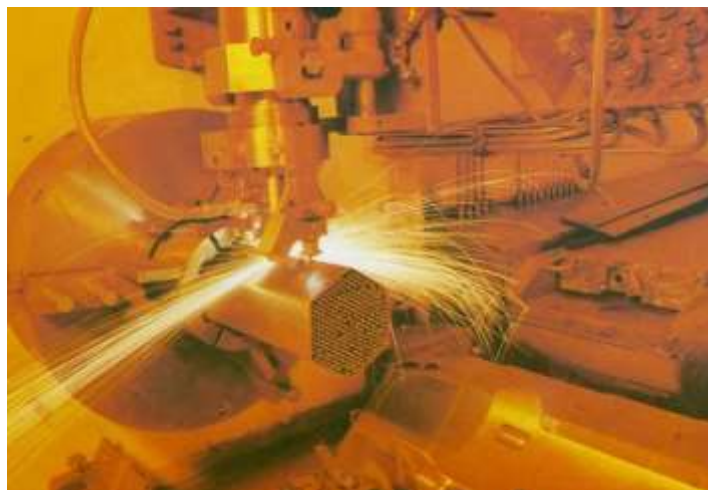


Figure 7 - Cutting of a FR fuel assembly

Fuel will be fed from the dismantling area to the shear pack equipment. Fuel will be cut into small pieces and fall via a chute into the pre-treatment equipment whilst off gas released will be routed to the dissolver off gas treatment system. A maintenance cell, located adjacent to the shear cell but separated using a shielded door, will enable the removal, replacement and maintenance of equipment via an MSM interface. Spares will be stored within this cell.

The throughput basis for this design is 5tHM/d, referring to the mass of heavy metal pre-irradiation. The approximate HM mass per FR fuel assembly (based on concept data) is 189.6 kg. Therefore to achieve the design throughput approximately 26.4 fuel assemblies would need to be fed into the system per 24 hours, equating to approximately 8726 fuel pins. This is considered a relatively high throughput, despite the fact that the plant would be operating 24 hours a day during campaigns, 200 days per year. Using THORP shearing operations as a basis, with an imposed maximum dissolution rate of 10 kgU/min [14], a shear-cave turntable capable of holding two fuel assemblies, and a maximum interruption time of 15.5 minutes (the time between shearing operations used to load assemblies from storage), it would take approximately 16 hours of continuous shearing operations to meet the design daily throughput. This basis would lead to an underestimate of time due to additional preparation tasks associated with the FR fuel. This could be shortened by the continuous dissolver as a higher dissolution rate could be imposed.

Further investigation into head-end throughput will need to be conducted at a later design stage. It has been assumed for the purpose of this design that the design throughput can be accommodated by a single mechanical preparation line with no parallel operations.

4.1.2. VOLOXIDATION

Chopped fuel pieces will feed via gravity into a two-stage voloxidation process performed within a single rotary kiln. As the kiln rotates the fuel will be contacted counter-currently with a pre-heated oxygen feed such that thermal and reagent gradients are established – precise control over the oxygen flowrate, temperature and kiln rotation is crucial and there may be some variance in process conditions required by the fuel. Trials will have to be conducted to establish the optimum process conditions for the feed however the kiln will be capable of reaching 1250 °C as a minimum.

The wall of the kiln towards the oxygen inlet can form a sieve through which pulverised fuel will pass to the dissolver (separating powder from the cladding hulls) however, to ensure all of the fuel is extracted from the hulls, there will be no separation of hulls from the fuel stream until the dissolver unit.

The feed to the rotary kiln is controlled by the rate of shearing, and thus the criticality safety within the kiln is dependent on a safe-by-mass approach, whereby only a given amount of fuel can occupy the kiln at any point in time. Voloxidation off gas will be routed to an off gas system separate to that of the dissolver. This system will be better suited to the removal of ^{85}Kr , ^3H , Cs , and ^{99}Tc whilst also removing ^{14}C and ^{129}I from the exhaust gas.

4.1.3. DISSOLUTION

Voloxidised fuel will be gravity fed via a chute into the charge hopper of a rotating wheel dissolver. This technology was selected due to its maturity; it has been operated at the La Hague reprocessing plants UP3 and UP2-800, since 1989 and 1994 respectively [15], and has the ability to finely control the rate of dissolution whilst maintaining criticality safety. It is believed that the reliability and experience of this type of dissolver offsets the advantages of other designs, despite the fact that some operate with no moving parts, decreasing maintenance requirements.

The dissolver will operate at 90°C and the fuel will dissolve into approximately 8M nitric acid before passing to the dissolver product liquor clarification units. Volatile fission products and dissolver off gas will be routed to the dissolver off gas system.

As the rotating wheel of the dissolver completes a rotation it will empty the cladding hulls into a chute. Hulls will be passed to a hulls packaging cell, where drums are filled with empty hulls, sealed and monitored to be transported and processed at a waste encapsulation plant.

A standby batch dissolver will be used to maintain dissolution capabilities should a problem with the rotary dissolver arise. This dissolver's primary role would be to allow for the dissolution of any chopped fuel which cannot be placed back into storage to make the plant safe should the primary dissolver fail. The design of this dissolver should mimic that used in THORP as a wealth of experience has been gained with this technology, though the higher Pu concentrations will need to be taken into account. A swing-style coupling similar to that used when switching between the three batch dissolvers in THORP will be adopted to divert material to the backup dissolver whilst maintenance and recovery operations take place.

Post dissolution, the liquor will be transferred to a NO_x conditioning column where it will be contacted counter-currently with nitrogen dioxide to ensure species are in the correct valence for extraction. Off gas will again be routed through the dissolver off gas system.

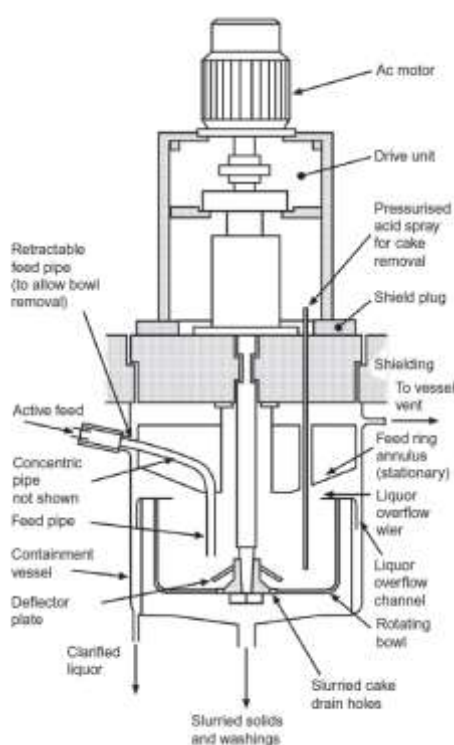


Figure 8 - Clarification Centrifuge [10]

The conditioned liquor will be continuously fed into a bowl centrifuge (Figure 8) (like those adopted on THORP). The rotating bowl will spin the liquid, forcing heavy particles to accumulate on the surface of the bowl. Periodically, the bowl is washed using pressurised nitric acid, with solids and insolubles (including insoluble Pu) washed out and eventually diverted to solid waste streams (after having first been treated to remove any Pu).

There is the potential for a criticality issue within the first centrifuge due to the expected levels of plutonium. The addition of neutron poisons, careful design of centrifuge geometry and improved acid dissolution could abate this problem however this will need to be addressed in more detail at later design stages.

Two Ag(II) dissolvers will be utilised to dissolve any insoluble Pu remaining after acid dissolution. These will each operate on a batch basis such that as one was undergoing dissolution the other would be filling with feed slurry from the washing of the first clarification centrifuge. Due to the relative maturity of the technology compared to ozonolysis, stirred electrochemical cells will be utilised adopting Ag(II) as a catalyst.

The dissolver will be constructed from corrosion resistant materials e.g. titanium with platinum electrodes. The equipment will operate at atmospheric pressure and 40°C.

There will be a common off-gas system for the two types of dissolver; this will be based on the design discussed in section 5.1.2.5.

The dissolution liquor from the Ag(II) dissolvers will undergo NO_x conditioning and clarification in a second centrifuge before passing to the aqueous mixer buffer tank, where it is mixed with the acid dissolver product liquor. The dissolution liquors are mixed and monitored in the criticality safe (by geometry) buffer tank prior to being sent to the chemical separation area of the plant.

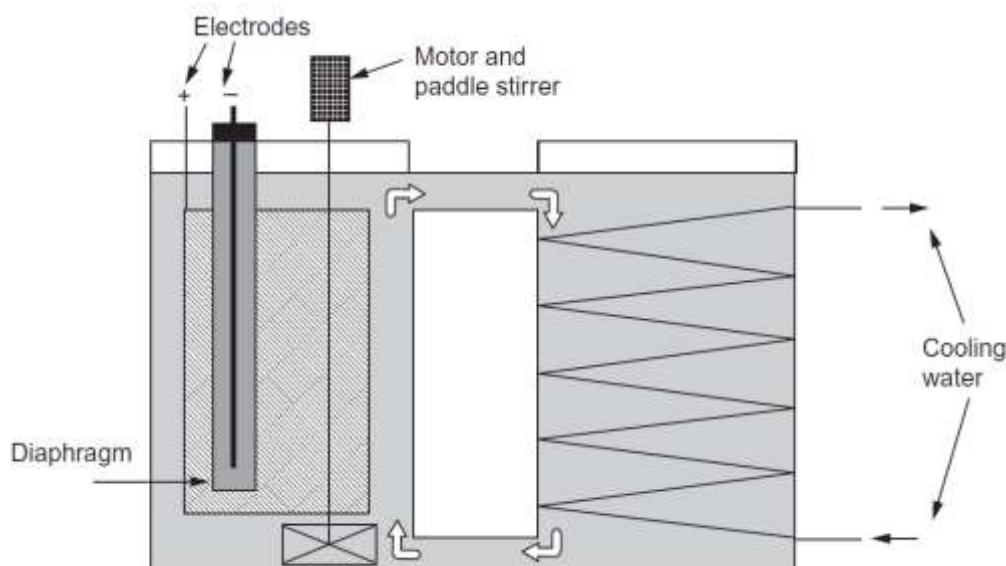


Figure 9 - Industrial plutonium dioxide dissolver [16]

4.2. CHEMICAL SEPARATIONS

4.2.1. EURO GANEX

Centrifugal contactors have been selected as the solvent extraction technology throughout the plant, due to the enhanced performance offered whilst occupying a relatively small footprint. In addition, this technology has been proven extensively in other industries and is increasingly being used in nuclear environments. All feeds to the centrifugal contactors will flow via constant head tanks, which adopt an overflow mechanism to ensure a constant head of liquid is fed to each contactor.

This section should be read in conjunction with Appendix 11.1 which details the results of flowsheeting exercises through summary diagrams

4.2.1.1. 1ST CYCLE

The purpose of the 1st cycle is to extract U from the FPs, actinides and lanthanides, which are then sent to the 2nd cycle.

The dissolution product liquor sent from the head end aqueous mixer and buffer tank will fill one of two metering tanks. These will be filled and emptied in turn to maintain a constant flow of feed to the chemical separation area of plant. Each tank will be sized at 35m³. This will account for one day's worth of material (33.11m³) plus contingency and will be geometrically safe.

The feed passes from the metering tanks to the **Primary Separation** bank of centrifugal contactors, comprising of 16 stages (individual contactors). The bank is split into two sections: contactors 1-10 form the extract section and contactors 11-16 form the scrubbing section. The feed enters the bank between stages 10 and 11. In the extract section feed is contacted with 1M N,N-di-2-ethylhexyl-isobutyramide (DEHiBA) dissolved in odourless kerosene (OK), which enters at stage 1. The DEHiBA selectively extracts the uranium and some actinide species (particularly technetium and neptunium) from the feed into the organic phase. In the scrub section the solvent is contacted with hydrazine to re-extract technetium and neptunium back to the aqueous phase.

Loaded solvent from stage 16 (**Primary Separation**) is transferred to the **Uranium Backwash** bank of centrifugal contactors. In this bank (comprising of 16 contactors) the solvent enters at stage 16 and is backwashed with 0.01M nitric acid, fed from stage 1, which strips the uranium back to the aqueous phase. The aqueous uranium stream leaves stage 16 and is sent to the uranium finishing area of the plant. Used solvent leaves stage 1 and undergoes a solvent wash before recycling back to the Primary Separation contactor bank.

The U-stripped aqueous stream (containing the bulk of the fission products, actinides and lanthanides) from the Primary Separation contactor bank leaves at stage 1 and is transferred to the Euro-GANEX 2nd cycle.

4.2.1.2. 2ND CYCLE

The 2nd cycle can be split into two sections: **Transuranic (TRU) Extraction and Scrub**, and **TRU Back Extraction**. In this cycle the fission products are first separated from the actinides and lanthanides, the actinides are then extracted followed by the lanthanides to form three output streams.

Prior to entering the TRU Extraction section, aqueous CDTA is added via a mixer to prevent the extraction of fission products (particularly zirconium and palladium) in the section.

The **TRU extraction and scrub** bank consists of 16 stages. Stages 1-12 make up the extract section where the feed (entering between stages 12 and 13) is contacted with 0.5 DMDOHEMA and 0.2M TODGA in OK entering through stage 1. The actinides and lanthanides are extracted into the organic phase, leaving the fission products in the aqueous phase which leaves the contactor at stage 1 and is taken off as waste. Stages 13-16 are used to scrub any remaining FPs from the actinide/lanthanide-rich solvent stream - 0.5M nitric acid is used to do this. The loaded solvent stream leaves the contactor via stage 16 and is routed to the **TRU Back Extraction** section.

The **TRU Back Extraction** section can be divided into three sub-sections: **Actinide Strip 1** (stages 1-6), **Actinide Strip 2** (stages 7-12) and **Lanthanide Strip** (stages 13-16). An/Ln-loaded solvent is fed between stages 6 and 7. Fresh solvent (0.5M DMDOHEMA and 0.2M TODGA) is added at stage 1. Strip acid (0.5M nitric acid, 0.055M BTP and 1M AHA) is fed between stages 12 and 13 (between the **Actinide Strip 2** and **Lanthanide Strip** sections). The acid is contacted with the solvent, extracting the actinides which leave as an aqueous product stream via stage 1 to go on to mixed actinide finishing. The solvent, which contains only bulk lanthanides, passes to stage 13 where it is contacted with 0.01M nitric acid (fed at stage 16). The remaining lanthanides are stripped and leave the bank at stage 13. Used solvent leaves via stage 16, undergoes a solvent wash and returns to both the **TRU extraction** and **Actinide Strip 1** sections.

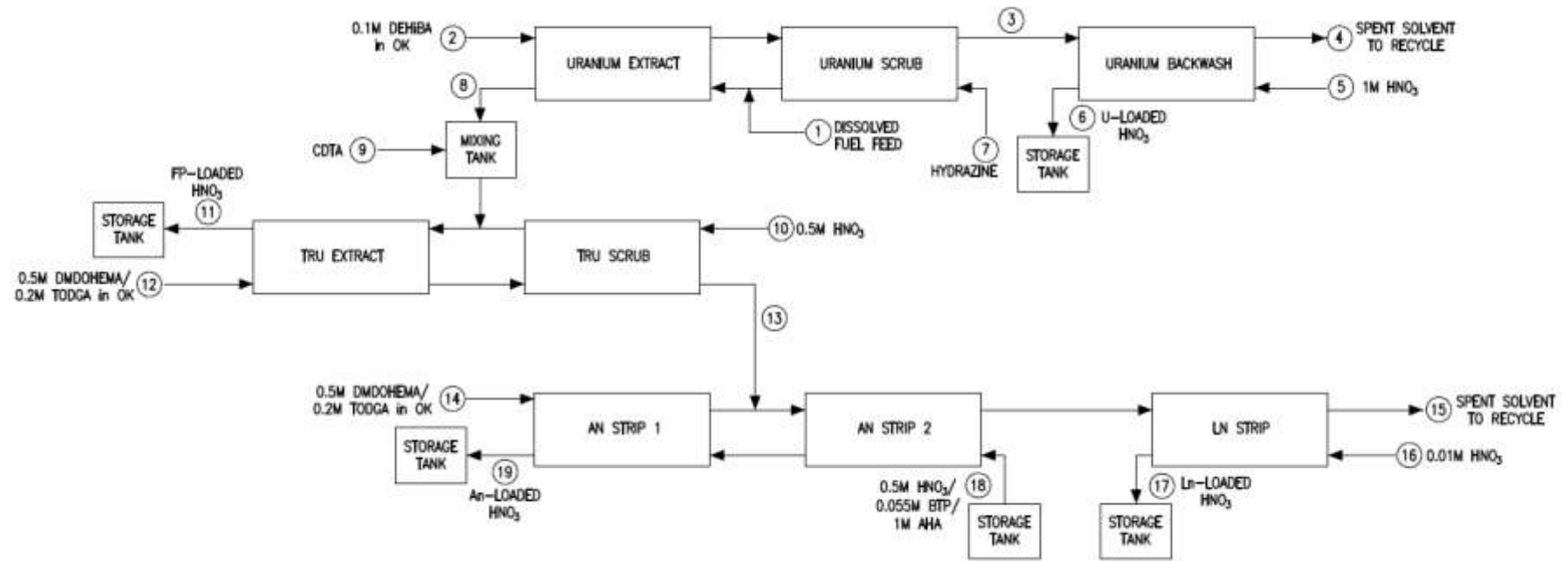


Figure 10 - Euro-GANEX Chemical Separation Flowsheet [2]

4.2.1.3. SOLVENT WASH

Over time it is expected that the solvent will degrade due to radiological damage. Solvent degradation products can impact of the effectiveness of the separation stages and, in some circumstances, present a hazard (material can be diverted to the wrong sections of plant which could result in a recycling of fissile material resulting in a criticality) [17]. It is desirable to wash and recycle solvent to remove degradation products, entrained FPs, actinides, and lanthanides. This would also minimise the quantity of spent solvent which is sent to HLW treatment, and thus minimise fresh solvent requirements.

To date there have been no studies into the solvent washing of the specific solvents used within the Euro-GANEX process. As a result, the solvent wash for this concept design shall be based upon the process adopted within THORP.

Within a centrifugal contactor bank (assumed to comprise of 16 stages per solvent wash – 32 in total) the solvent will be contacted with 0.1M nitric acid and 0.05M hydroxylamine (N_2OH) to reduce any Pu(IV) to Pu(III), and Np(VI) to Np(V). These are less soluble in the organic phase and are backwashed into the aqueous phase.

The solvent then passes to another contactor bank where it is contacted with 0.25M sodium carbonate (Na_2CO_3) to remove any acidic solvent degradation product that would affect plant performance. Np and Ru salts are also removed with the aqueous phase.

The solvent is then contacted with 0.01M nitric acid to remove any remaining alkali in the solvent, and acidifies the solvent for re-use.

Washed solvent is stored in a buffer tank to decouple it from the process. Here the solvent can be sampled and monitored, spent solvent can be purged from the process and fresh solvent added. It is assumed that the spent solvent will be compatible with existing spent solvent waste treatment processing technologies.

To flowsheet the two solvent recycle loops (see Figure 22) a “feed and bleed” approach is adopted whereby some spent solvent is purged and the remaining solvent is topped up with fresh solvent before returning to the system. Though the flowrates of solvent to and from the system may not be representative of reality (which would be more conservative) this method does model the removal of some dissolved FPs, An and Ln representing the washing cycles.

4.2.1.4. SILVER RECOVERY

To recover reusable, expensive silver from the aqueous FP waste stream two electroplating units will be employed operating such that as one is being filled the other is operating and emptying. The feed will be conditioned with OK within a centrifugal contactor bank (assumed to consist of 8 stages) to remove any entrained solvent. The liquor will then pass through one of the two batch electroplating units. Once a batch is complete the electrodes are washed using nitric acid to remove solids. The electroplating vessel is then heated using electrical heaters and the silver is dissolved in 4.5M nitric acid for recycle to the silver dissolver.

4.2.1.5. CONTACTOR SIZING

Table 1 below illustrates the number of contactors proposed for each chemical separation section and the rotor diameter.

Table 1 - Calculated Centrifugal Contactor Sizes

Unit	total feed (l/min)	Contactor size (cm) - assumes 50% hydraulic limit	Number of contactors in section
Primary Separation - Extract Section	35.61	20.81	10
Primary Separation - Scrub Section	88.31	29.73	6
Uranium Backwash	153.29	36.93	16
Extract 1 & 2 (2nd Cycle Extract)	133.64	34.99	12
Scrub 1 & 2 (2nd Cycle Scrub)	92.22	30.24	4
An Strip 1	123.96	33.97	6
An Strip 2	170.52	38.51	6
Ln Strip	176.13	39.00	4

4.3. URANIUM FINISHING

In principle, the uranium finishing section of plant must concentrate the aqueous uranyl nitrate from the U backwash to approximately 1000g/l by evaporation. Then Thermal DeNitration (TDN) will convert the concentrated stream to a uranium trioxide product, ready for packaging and storage in 50L drums. It is envisaged that the finishing equipment will either be contained within large gloveboxes or cells spanning several floors to utilise gravity to transfer liquors to subsequent process steps where possible.

Two buffer storage and preparation tanks will receive aqueous uranyl nitrate (UN) from the uranium backwash contactor bank. These will each hold a day's worth of material ($112.5\text{m}^3/\text{d}$) and fill in turn as the other passes its material to the next process unit. The tanks will be geometrically safe for criticality safety. The liquor will be monitored to check its enrichment level, with samples taken to confirm these measurements, as the enrichment must be sufficiently low to prevent criticality problems downstream. Down-blending with low enriched UN, if required, can be performed in the storage and preparation tanks. A feed preparation tank is required to prepare the low enriched Uranyl Nitrate (UN) feed on a batch-wise basis. Solvent will accumulate at the surface of the prep tanks and thus a solvent float off system will be adopted to take off any solvent entrained with the aqueous feed. This solvent will be treated with the waste solvent from the chemical separation area.

Prior to passing to the evaporator, the feed passes through a stream-strip column to destroy any remaining solvent. A vertical thermosiphon calandria evaporator, like those adopted in THORP, will be used. This will operate at approximately 90°C at a reduced pressure, with concentrate continuously withdrawn to a concentrate receiver. The crystallisation temperature of the concentrate is approximately 60°C therefore all further pipes and equipment are trace heated to above approximately 90°C . Two geometrically safe constant feeder tanks, each holding a day's worth of concentrate act as a break between the evaporator and the TDN reactor. Evaporator condensate is taken off as waste and sent to the appropriate facility on the site.

Prior to thermal denitration, sulphuric acid is added to the feed which increases the reactivity of the UO_3 for further processing.

The TDN reactor will be of a fluidised bed design operating at 300°C (by electrical heaters), with the UN feed being sprayed on to a bed of UO_3 powder fluidised by air. Excess UO_3 powder will over-flow from the reactor, undergo cooling by a stream of cool air, and pass into storage hoppers ready for sampling and drum filling. Off-spec powder can be recycled and fed back into the TDN reactor feed tanks. To do this a small dissolver vessel enables the powder to be re-dissolved into UN form. The reactor off gas is cooled and filtered before passing through a caustic scrubber (to remove NO_x gases) and to the building stack.

Product powder drums are stored prior to being exported to a dedicated product storage building.

4.4. MIXED ACTINIDE FINISHING

The mixed actinide finishing section of the plant is expected to mimic that of the plutonium finishing line of THORP. Similar to U finishing, the mixed actinide finishing is expected to occur in either large gloveboxes or cells. The addition of a solvent destruction and evaporation stages have been deemed necessary due to the more dilute stream (relative to THORP) expected to arise from the chemical separation area.

Actinide-bearing aqueous liquor will be homogenised and sampled in product storage buffer tank before leaving the chemical separation area. The liquor is then transferred via reverse flow diverters (RFD's) to the steam strip column which destroys any entrained organics before passing into the evaporator.

The evaporator is expected to be of a similar design to that used in the uranium finishing section however care will have to be taken to ensure that the evaporator remains safe in terms of criticality geometries, and that the evaporation does not produce an over-concentrated product which in turn would present a criticality hazard. The concentrated liquor will be metered via continuous volume feeders (CVFs) into a conditioning vessel where hydrogen peroxide is added to modify the valence of the actinides such that the crystals produced during precipitation will be of the correct size and shape.

In THORP, two dilution vessels add nitric acid to the feed to reduce the concentration of plutonium for the precipitation stage. It is expected that these units will not be necessary due to the dilute plutonium concentration in the feed from chemical separation – the concentration of plutonium will be controlled by the extent of evaporation. Trace heated pipes pre-heat and carry the liquor, via a CVF, to precipitation.

Liquor is continuously fed into the precipitation vessels along with pre-heated oxalic acid (four vessels are used in parallel in THORP, it is assumed this will be sufficient capacity for this plant). The oxalic acid reacts to form plutonium oxalate precipitate at approximately 60°C. The vessels are stirred such that a vortex is formed, the precipitate slurry overflows from the lip of the vortex and falls on to a flat-bed rotary vacuum filter. The slurry is vacuum dried and the filter cake washed with dilute nitric acid and demineralised water before being removed using a screw feeder. The moist cake is removed to the drying furnace. The remaining oxalate mother liquor (OML) is removed and evaporated to form a concentrated mother liquor (CML) which in turn undergoes evaporation again to destroy residual oxalic acid, it is then stored prior to re-use within the process.

The plutonium oxalate is fed through the drying furnace using rotating screws against a contra-flow of process air and argon. The off gas is passed through a filtration unit to remove particulates before passing to the glovebox ventilation system. The drying process converts plutonium oxalate to dioxide powder which is then fed into a calcination furnace for surface area adjustment using process argon.

Product powder flows into a hopper and feed tube to the batch blending unit, which homogenises the powder prior to sampling and packaging. Blended powder fills a storage hopper for sampling prior to storage in triple-

packaged stainless steel cans, each with bar-code identification. The cans are monitored for surface contamination and taken to a buffer storage area before being exported to the product store. All product storage contains active (and backup passive) cooling facilities with criticality-safe racking.

4.5. KEY EQUIPMENT LIST

The table below lists the key plant items required, this is indicative of the main unit operations and is by no means exhaustive.

Table 2 - Key Plant Equipment

Plant Area	Plant Item	Number off
Head End	Shear Machine	1 Off Duty
	Rotary Kiln Voloxidiser	1 Off Duty
	Rotary Wheel Dissolver	1 Off Duty
	Batch Dissolver	1 Off Standby
	Ag Dissolver	1 Off Duty 1 Off Standby
	NO _x Conditioning Column	2 Off Duty
	Centrifuge	2 Off Duty
	Accountancy Tank	1 Off Duty
Chemical Separation	Metering Tanks	1 Off Duty 1 Off Standby
	Centrifugal Contactors	32 Off Duty (Solvent wash) 64 Off Duty (Euro-GANEX) 8 Off Duty (Silver Recovery) 16 Off Standby
	Silver Recovery Unit	1 Off Duty 1 Off Standby
U Finishing	Buffer Storage Tanks	1 Off Duty 1 Off Standby
	Feed Preparation Tank	1 Off Standby
	Steam Strip Column	1 Off Duty
	Evaporator & Condenser	1 Off Duty
	Constant Feeder Tank	1 Off Duty 1 Off Standby
	Thermal Denitration Reactor	1 Off Duty
MA Finishing	Buffer Storage Tanks	1 Off Duty 1 Off Standby
	Steam Strip Column	1 Off Duty
	Valency conditioning tank	1 Off Duty
	Evaporator & Condenser	1 Off Duty
	Precipitation Vessels	4 Off Duty
	Rotary Vacuum Filter	1 Off Duty
	Calciner	1 Off Duty
	Blending Unit	1 Off Duty

5. TECHNICAL OPTIONEERING

A technical assessment for each of the major unit operations within the process outlined by Figure 2 has been undertaken as part of this design.

5.1. HEAD-END OPERATIONS

5.1.1. PRE-TREATMENT OF FUEL

5.1.1.1. MECHANICAL PREPARATION

5.1.1.1.1. FUEL SHEARING

The chemical separation of cladding from fuel is not appropriate for fast reactor fuel as the stainless steel cladding is difficult to dissolve using standard reagents [10]. As a result, mechanical shearing is required to expose the oxide fuel for dissolution.

Feed fuel is assumed to be stored underwater in MEBs suitable for the fuel type within the receipt and storage pond. Fuel assemblies will be removed from the MEBs and transported via an elevator to the mechanical shearing equipment, located so that the sheared fuel will feed the pre-treatment and dissolver equipment via gravity. Off-gas released upon the breaking of the fuel pin containment will be routed to the dissolver off-gas system. The fuel is aligned with the shearing pack envelope and forced a sufficient distance past the chopping block by a hydraulic ram to obtain fuel pieces of an optimum size (50-100mm are typical) [10]. The fuel is then chopped by a hydraulic guillotine blade and fed to the next pre-treatment stage.

Mechanical shearing is widely used and is technically mature. For this reason, it is assumed that the guillotine style equipment adopted by THORP (shear pack), made suitable for the FR fuel will be used. This equipment is modular in design allowing for maintenance and replacement via an adjacent cell and MSM.

The geometry of fast reactor fuel is expected to vary from traditional thermal fuel assemblies. The pins are expected to be thinner in diameter which could cause crimping issues if the assembly was sheared as a whole assembly (as thermal fuel is sheared in THORP). Crimping is not an issue for larger pins as good access to the oxide within the fuel pieces is still available. However, crimping could significantly restrict access to the oxide in fast reactor fuel hindering further pre-treatment and dissolution operations [10]. It has been recommended that fast reactor fuel assemblies be dismantled and either sheared as single pins or as linear arrays of pins to minimise crimping effects [10]. The throughput of a full scale reprocessing plant may require an approach whereby fuel is disassembled and stored in appropriate fuel pin racks prior to operational campaigns.

Precise control over the hydraulic ram and blade rates are required as these will largely determine the feed rate of fuel into the process.

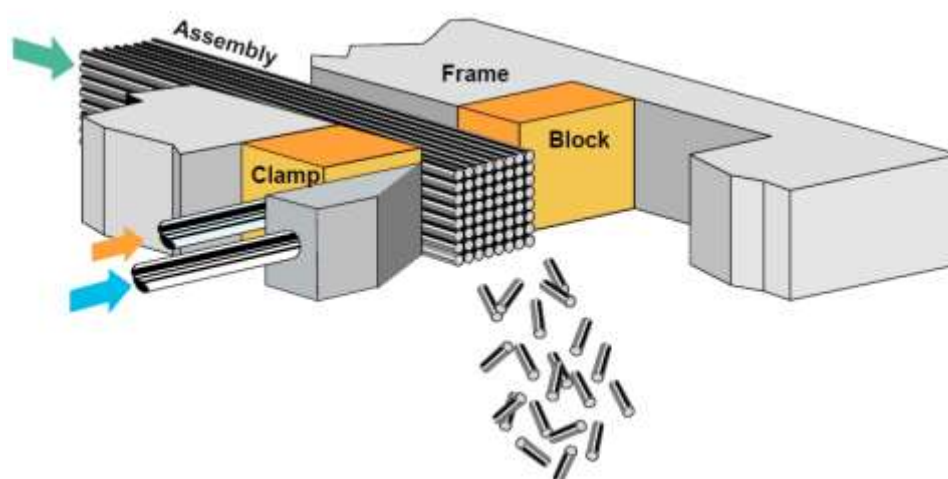


Figure 11 - Schematic of shearing device [18]

5.1.1.1.2. FUEL PUNCHING

Oxidation processes, such as volatile oxidation (voloxidation) (see section 5.1.1.2), rely upon exposure of the oxide fuel to the atmosphere within the furnace(s). Fuel punching is an alternative to mechanical fuel shearing which involves puncturing the cladding of a fuel pin along its length using a mechanical device operated by a MSM, providing access to the fuel surface area for the oxidative process to occur. This method relies entirely on the oxidative process to release the fuel from the cladding, and as such there is less control over the size and shape of cladding waste generated. Though this method may be well suited for small voloxidation and dissolver throughputs, it may struggle with the high throughput of 5tHM/d. In addition the voloxidation furnaces would have to be designed to accommodate a whole fuel pin. Therefore shearing will be chosen over punching for this design.

Alternative oxidation processes such as Oxidation Reduction Oxidation (OREOX) and Atomics International Reduction Oxidation (AIROX) could also be used with this technology.

5.1.1.2. VOLOXIDATION

Voloxidation is a high-temperature pre-treatment process for spent nuclear fuel (SNF) which can be employed prior to dissolution in nitric acid. Voloxidation generally enhances the dissolution rate of the fuel, however it also increases the amount of Pu that remains in the undissolved residues, which can cause problems for downstream units [10].

Sheared oxide SNF is oxidised from UO_2 to U_3O_8 by thermal treatment using oxygen in a furnace or rotary kiln. The fuel pulverises due to a change in crystalline structure which increases its volume. As a result the cladding is placed under pressure and cracks, releasing the fuel. The product is sieved to remove cladding, though a small amount of fuel is entrained with the cladding. Other actinides cannot be further oxidised and remain in the dioxide form.



Figure 12 - Spent Nuclear Fuel Post Voloxidation [19]

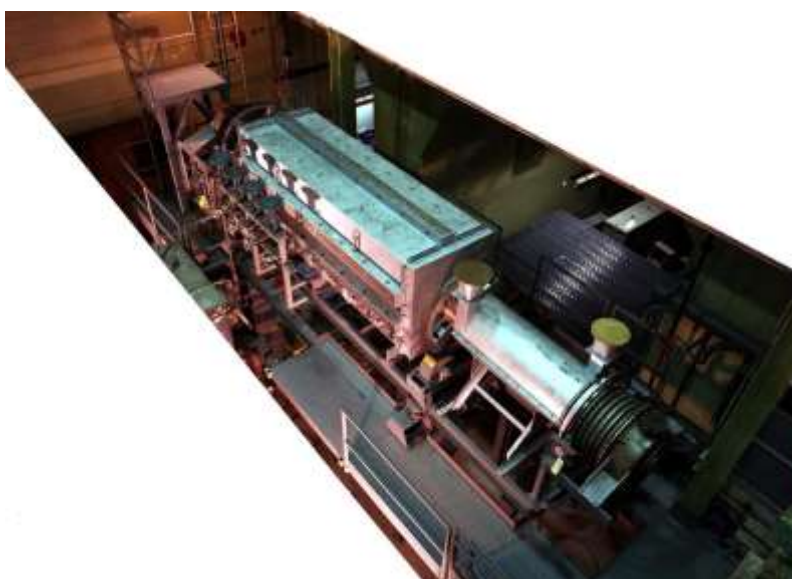


Figure 13 - Rotary Kiln Used for Voloxidation [20]

Voloxidation can occur over a large temperature range, with higher operating temperatures giving rise to increased release of volatile fission products; conversely high temperatures can lead to Pu sintering. Further development work is required prior to the incorporation of voloxidation into a full scale reprocessing plant, notably regarding the pulverisation of MOX powders with relatively high Pu concentrations.

Voloxidation can be performed as either a single oxidation or two-step oxidation/reduction process. Both single and two-step processes produce a pulverised powder product, the key difference is the oxidation state of the product. The addition of a reduction step has been suggested to increase compatibility with traditional dissolution technologies; reduction creates a dioxide product, the form in which most commercial dissolution is undertaken, hence there is a higher technical maturity associated with it. On the other hand, the dissolution of fuel directly from a single oxidation stage would be relatively straight-forward.

The oxidation/reduction has a two-fold pulverisation effect on the fuel, therefore there should be a greater separation of fuel from cladding and less fuel lost to waste. However, due to the fact that oxidation and

reduction should take place in separate furnaces, there is a larger demand for equipment and space with the two-step process, but a single furnace could be adopted if a temperature and reagent gradient was maintained.

Voloxidation and dissolution can present many advantages compared to traditional direct dissolution methods such as:

- The density decrease associated with oxidation causes the fuel to swell and pulverises itself into a powder. This releases the fuel from the cladding and increases the surface area available for dissolution.
- Volatile fission products such as ^{14}C , ^3H and ^{129}I , are released as gases and can be treated prior to dissolution.
- The volumetric flow of HNO_3 required for dissolution can be reduced as UO_2 is converted to U_3O_8 . Volumetric flow of dissolver off-gas is also reduced; however voloxidation off gas must be treated separately.

A rotary kiln would typically be used for voloxidation in a continuous process. This would comprise of a rotating cylindrical vessel heated using either a furnace or electrical heaters. This technology is well developed in other industries, as well as in the treatment of high level radioactive waste through vitrification. The kiln must be capable of reaching a minimum temperature of 1250 °C, the actual operating temperature will have to be determined through research and development. The powder product can pass through a sieve where large pieces of cladding can be removed and monitored.

Voloxidation is expected to release a number of off-gases including: ^{14}C , ^{85}Kr , ^{129}I , ^3H , Cs, and ^{99}Tc . These will have to be treated via a separate off gas system to the dissolver. The Korean Atomic Energy Research Institute (KAERI) has developed such an abatement system tested by themselves and the Idaho National Laboratory (INL) [21]. The system consists of a fly ash filter to trap Cs, a calcium-based ash filter for Tc and a silver silicate filter for the trapping of iodine. ^3H and ^{14}C are also trapped in each filter, and ^{85}Kr emission is controlled via the gas passing through a decay tank.

Voloxidation will be incorporated into this design due to the increased dissolution rate and reduced loss of fuel to waste (with the cladding).

5.1.2. FUEL DISSOLUTION

Fuel which contains low concentrations of Pu (<15% w/w) can be dissolved via direct dissolution with nitric acid. This is a mature technology which is employed in commercial reprocessing plants.

Fuel containing high concentrations of Pu (>15% w/w) such as fast reactor fuels may incur problems with direct dissolution in nitric acid. A small percentage of the plutonium may be insoluble in the nitric acid. In addition, the reaction mechanics of dissolution can become more dependent on the concentration of Pu than the concentration of U. As a result, insoluble Pu has the potential to enter the solvent extraction cycles, recycle within the system and accumulate, presenting a criticality risk. To prevent such a hazard, an enhanced dissolution stage would be required to treat the insoluble particles.

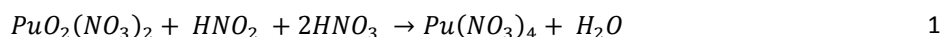
The quantity of Pu-rich fines is dependent on the nature of the fuel, its burnup and the initial concentration of Pu pre-irradiation; however quantities tend to increase with burnup and Pu concentration. Fines can contribute as much as 0.74% w/w in FR fuel [10]. The insoluble Pu remains proliferation resistant due to the impurities within the process streams.

The design of dissolver equipment for FR SNF is made more difficult, relative to thermal SNF, due to the increased Pu concentration as more care needs to be taken with regard to criticality prevention and shielding.

Post dissolution the dissolver liquor is conditioned in preparation for the chemical separation section of plant. This is to:

- Remove iodine
- Ensure that the acidity and the valences of U and Pu are acceptable for solvent extraction.
- Remove solid particles.

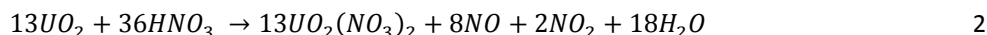
Both the removal of iodine and valence conditioning can be achieved using an air and nitrogen dioxide sparge. Batch dissolvers, like those employed in THORP, perform this sparge at the end of dissolution. A sparge is not desirable in continuous dissolvers due to the relatively slow rate of dissolution. For this reason a separate NO_x conditioning stage is recommended. The liquor is typically contacted with the gas mixture counter currently in a column. Equation 1 [10] outlines the reduction of Pu to the tetravalent state (preferred for solvent extraction processes).



The removal of solids is achieved using a centrifuge. The centrifuge operates via a continuous feed with a periodic discharge of slurried-solids and washings. As a result the geometry must be carefully considered to conform to safe-by-shape geometry to avoid a criticality risk. In practise two centrifuges would operate together: whilst one was being fed with liquor and operating, the other would undergo a wash-out procedure.

5.1.2.1. DISSOLUTION OF SHEARED FUEL - WITHOUT VOLOXIDATION

Direct dissolution is typically used to dissolve fuel in the form of UO₂. Equation 2 illustrates the hybrid reaction that occurs.



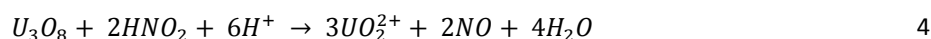
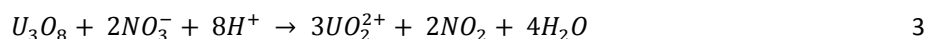
Almost all (95.5-100%) of the Pu contained within the fuel will dissolve, though the exact amount is dependent on the fuel fabrication method and concentration of Pu within the fuel. As mentioned above, high Pu concentrations will produce a larger quantity of insoluble particles, requiring additional treatment of the solution such as enhanced dissolution techniques. Direct nitric acid dissolution technology is very effective for the dissolution of U oxides, however it is much less effective for other actinides such as PuO₂ and NpO₂. The dissolution of MOX fuels can become problematic due to the inherently higher Pu concentrations which, as described above, can present criticality issues. Similarly spent fast reactor fuel is also problematic due to its increased plutonium concentrations relative to thermal oxide fuels.

Fission products adopt the forms M, M⁺, M²⁺, and M³⁺. These react in a similar manner to U forming nitrates and water in the dissolver.

In the absence of voloxidation most fission product gases must be handled within the acid dissolver. Iodine can dissolve into water via a reversible reaction, however this is mostly removed from the solution in the NO_x conditioning stage. All off gas is routed to the dissolver off gas treatment in this scenario.

5.1.2.2. DISSOLUTION OF VOLOXIDISED FUEL

Depending on the mode of voloxidation operation adopted either a UO_2 or U_3O_8 product will feed into the dissolver. If a two-stage oxidation/reduction method is adopted, a UO_2 product will form and dissolution will occur as in Section 5.1.2.1. Equations 3 and 4 illustrate the dissolution of U_3O_8 formed by a single oxidation stage.



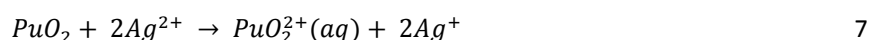
5.1.2.3. ENHANCED DISSOLUTION

The higher quantities of Pu in FR fuel relative to TR fuel mean that the likelihood of undissolved Pu in the product liquor of a standard direct dissolver is increased. This can result in a recycle of fissile material if allowed to proceed to chemical separation, presenting a criticality hazard. Enhanced dissolution must therefore be used to ensure as much Pu as possible is dissolved prior to chemical separations. There are three known methods for the dissolution of Pu oxide [10]:

- Dissolution using strong complexants e.g. fluorides
- Oxidative dissolution e.g. using Ce(IV) or Ag(II)
- Reductive dissolution e.g. using Ce(II) or U(IV)

The first method raises issues with corrosion for the dissolver and downstream units. In addition the introduction of salts increases the volume of HLW produced. Similarly, there has been little research performed on reductive dissolution to date. Therefore, oxidative dissolution is the most mature enhanced dissolution technology for spent fast reactor MOX fuel.

Oxidative dissolution involves the use of a catalyst (mediator) and either an electrochemical or ozonolysis process to dissolve the plutonium and regenerate the mediator, and can be performed at ambient temperatures. Ag(II) or Ce(IV) can be used, but silver is preferred due to its higher oxidation potential, rapid electron transfer rate and stability against water oxidation. The mediator is generated at the anode of an electrochemical cell whilst nitric acid is reduced at the cathode, the mediator then catalyses the dissolution of plutonium into the anolyte (see equations 5 to 7) [10]. Dissolution is achieved in an electrochemical cell comprising of two compartments separated by a membrane which allows a transfer of protons across it but separates the anolyte and catholyte to avoid the reduction of mediator by the acid reduction products. This process is generally termed a Mediated Electrochemical Oxidation (MEO) process.



A MEO process is most likely to be utilised after direct dissolution (to further treat insoluble particles), mostly due to the ease, speed, and high technical maturity of direct dissolution. Ideally, it would take place in the same dissolver as direct dissolution as this would minimise the transport and risk of accumulation of Pu-rich solids however this would only be able to be accommodated in a batch process. Additionally, a single dissolver strategy would reduce the requirement for processing equipment and as a result may decrease capital costs

however there is the potential for additional side reactions to occur in addition to increased dissolver wear due to higher acidity requirements.

Continuous processes would require a separate process step, and care would need to be taken when designing this section of plant such that the transport of Pu-rich solids does not present a significant hazard. The two dissolver options would require additional equipment however it would allow the design of, and the conditions within, the second dissolver to be optimised. There is currently no design of a continuous electrochemical cell dissolver and as such to incorporate this technology into a continuous process a number of cells would need to be used in parallel. These would operate in such a way that whilst one was being fed with slurry another would be performing dissolution, monitoring solids content and emptying. Additional spare capacity may need to be considered.

There is little information available regarding the scale-up of enhanced dissolution processes, and for the purpose of this design an electrochemical cell may be assumed for this process. However in practise it would be undesirable to have electrochemical equipment in a high active (HA) environment given the continual maintenance required. One suggested alternative to this technology is ozonolysis using an ozone sparge as an oxidising agent, which could easily be incorporated into a continuous unit.

5.1.2.4. DISSOLVER DESIGN OPTIONS

Batch dissolution has traditionally been used in commercial reprocessing plants, though a small number of continuous concepts have been developed. In THORP, fuel is chopped and falls into the dissolver via a chute. Dissolution occurs over the course of several hours after which the solution is sparged with an air and NO₂ mixture and the baskets containing cladding are removed and sent to waste. Neutron poisons such as gadolinium nitrate can be added to the liquor prior to discharge to the next processing stage to reduce the risk of a criticality hazard.

Continuous dissolution offers several benefits when compared to batch dissolution. Remote manual handling can be significantly reduced in a continuous system as the need to move baskets around is eradicated. The flow of feed solution to the solvent extraction cycles can be better controlled in a continuous system, allowing for optimum extraction conditions. Continuous operation allows for the design of equipment that can both handle the high flow rates of a full scale plant whilst also being safe-by-shape in terms of geometry to prevent criticality.

A brief overview of the two main types of continuous dissolver design is provided below.

5.1.2.4.1. CONTINUOUS ROTARY DISSOLVER

There are two further types of continuous rotary dissolver. The first, developed by COGEMA [22], adopts a vertical wheel/carousel that has been divided into perforated radial sections. The wheel is suspended above a bath of nitric acid such that the lower sections (where dissolution takes place) are fully immersed in the acid. The inward faces of the sections are open such that fuel can be loaded via a chute into sections just above the acid surface, and hulls are tipped into a waste chute as the wheel rotates. The speed of rotation is such that the fuel in a section should be dissolved by the time it is raised from the acid. Both the compartments and acid bath are sized to minimise the risk of criticality. Figure 14 illustrates this design.

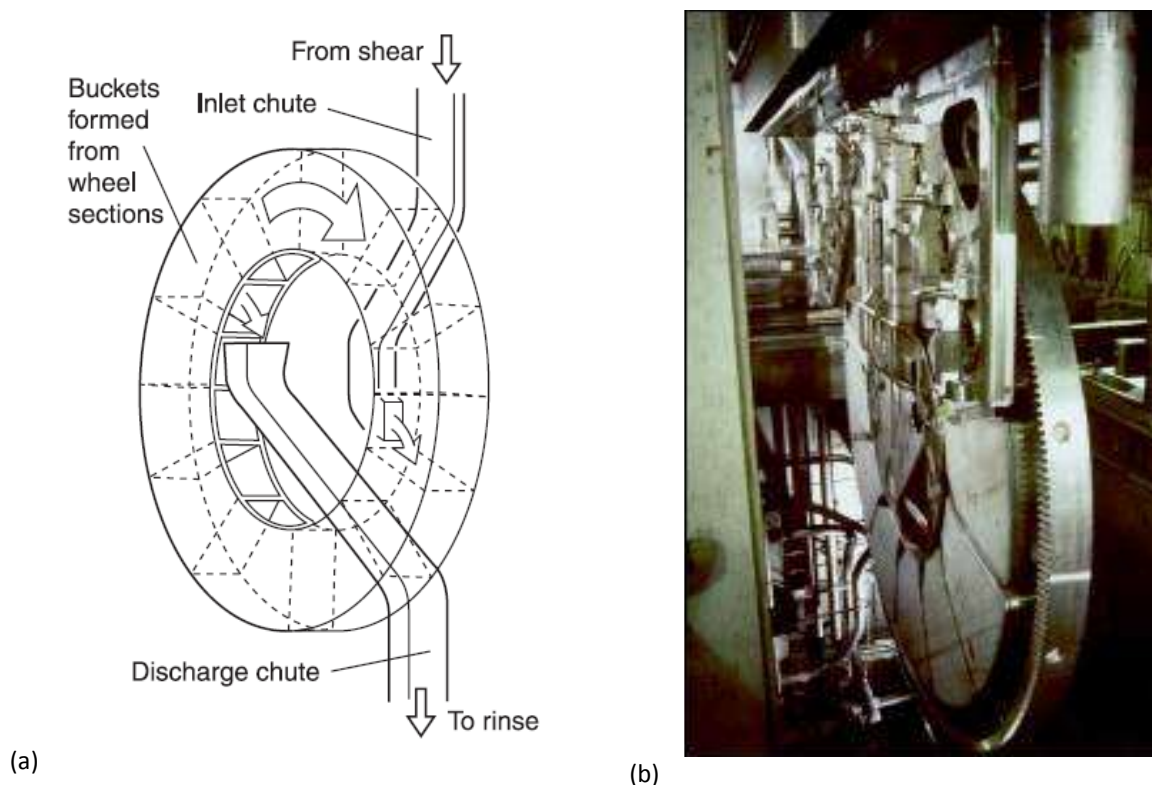


Figure 14 – Continuous rotary wheel type dissolver (a) Schematic [23], (b) UP3 dissolver [24]

Another type of continuous rotary dissolver resembles that of a helical screw (Figure 15). Fuel enters the dissolver via a chute and is gradually pushed horizontally along the dissolver whilst being contacted counter-currently with nitric acid. When the hulls reach the end of the dissolver they are expelled via a solid waste chute with dissolver product liquor exiting at the opposite end. The screw is designed such that the gaps between adjacent threads essentially form compartments designed to prevent criticality. In addition the spacing between compartments is sized to prevent criticality, though the feed rate to the dissolver could also control how “spread out” fuel is. A motor is used to turn and rock the screw such that the fuel is completely dissolved by the time the hulls reach the end of the dissolver.

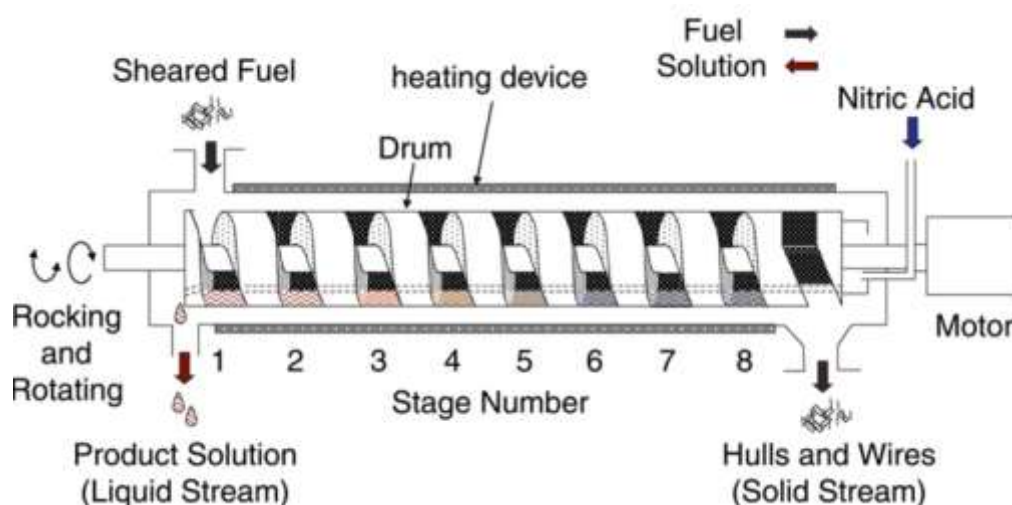


Figure 15 - Continuous rotary drum dissolver [25]

5.1.2.4.2. PULSED DISSOLVER

Pulsed dissolvers utilise pneumatic pulses as opposed to mechanical moving parts to move fuel through the dissolver. The simplicity and lack of moving parts reduces the need for maintenance and failures.

One design that utilises pulsing technology is the helical ramp dissolver (Figure 16). This consists of two concentric columns – the central column contains nitric acid, the annular space between the inner and outer column walls houses a helical ramp. Nitric acid is forced from the central column into the outer column at its base with each pulse. Sheared fuel is transported up the ramp with each pulse as the fuel is dissolved (the ramp is ridged to allow the fuel to come to rest in between pulses). As fuel dissolves the hulls become lighter making it easier for them to travel to the top of the equipment.

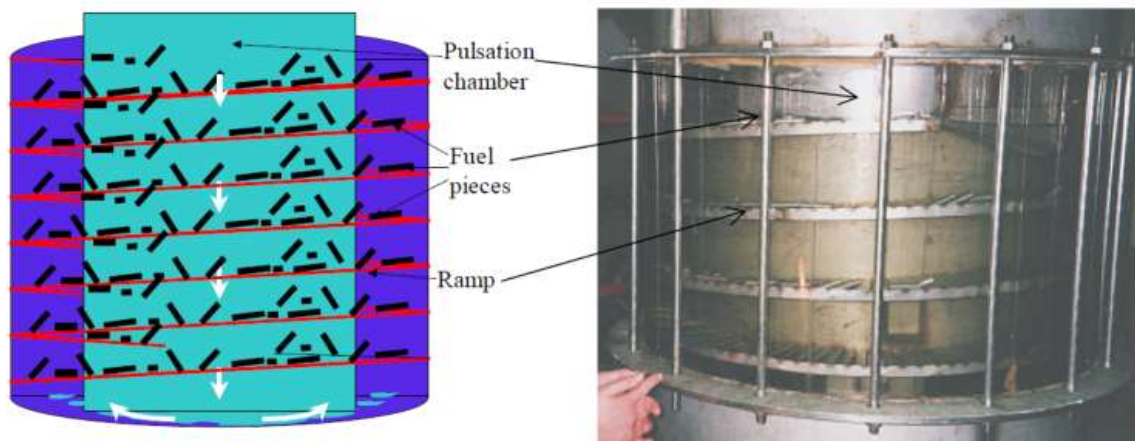


Figure 16 - Helical ramp pulsed dissolver [26]

A pulsed-V dissolver is another example of a pulsed dissolver. It utilises a V-shape to create a bath of liquor which completely immerses the sheared fuel pieces. Agitation is provided via pneumatic pulses which aids fuel dissolution. As the fuel dissolves and the hulls become lighter they overflow out of the dissolver. Due to the relatively small scale of this dissolver it is expected that multiples of them would be connected in series, each dissolving more fuel from the hulls. No evidence has been found supporting the implementation of pulsed-V dissolvers at an industrial scale.

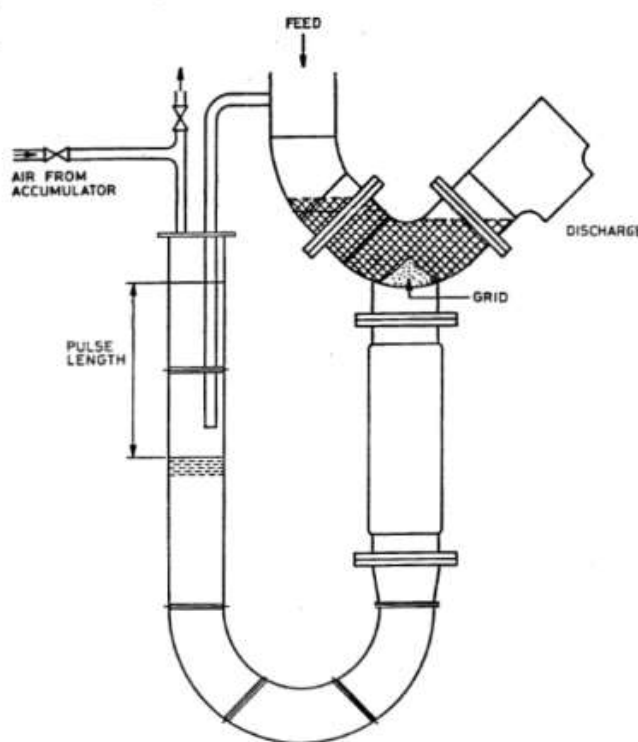


Figure 17 – Pulsed-V dissolver [27]

5.1.2.5. OFF GAS TREATMENT

The overall requirements of the dissolver off gas treatment are not expected to vary significantly to that used in thermal fuel reprocessing plants. The main difference is expected to be the incorporation of a separate off gas treatment stream if voloxidation is adopted.

It is expected that, if secondary dissolvers are adopted for enhanced dissolution they will share an off gas treatment system with the primary direct dissolver. The off gas system described below outlines the systems adopted in THORP.

The main objective of the off gas treatment process is to ensure that the off gas does not exceed the radioactive and chemical discharge limits of the site. Species of concern are:

Active

- ^{129}I In the form of I_2
- ^{14}C In the form of $^{14}\text{CO}_2$
- ^{106}Ru In the form of RuO_4 and RuO_2
- FPs In the form of fuel dust particles

Non-Active

- Nitrogen Oxides (NO_x)

Off gas abatement will generate some effluent wastes. These are expected to take the form of recombined nitric acid, spent caustic and weak acid.

A high flow rate of air must be induced through the off gas system to ensure fine particles from shearing operations are drawn into the dissolver and not into the shearing cave/area (a continuous negative pressure must be maintained). This has the downside of significantly increasing flowrate requirements and diluting the species to be removed from the gas.

The stages within the THORP dissolver off gas treatment process are outlined in brief below.

1. Condensation
 - Condenses and returns water vapour back to the dissolver.
 - Fuel dust is removed.
 - Significant levels of NO_x can be absorbed in the condensed steam.
2. Acid scrubbing column (HNO_3)
 - Removes NO_x , fuel dust particles, some I and some Ru.
3. Iodine desorption column
 - Removes iodine from the spent acid from the acid scrubbing column by contacting it with air.
4. Plug flow reactor
 - Oxidises NO to extractable NO_2 in the acid scrubbing column product in preparation for the caustic scrubbing column.
5. Caustic scrubber
 - Utilises NaOH to remove NO_x , ^{129}I and ^{14}C to acceptable discharge levels.
 - Some Ru is also removed.
6. Weak acid column
 - Removes entrained NaOH and dehumidifies the gas in preparation for filtration.
 - Some Ru is also removed.
7. Filtration
 - HEPA filters are adopted as a last barrier to trap any active material prior to the discharge of the gas.
 - The filters can be centralised for the entire reprocessing plant. Care must be taken to ensure the off gas feeds meet the process requirements of the filters.

5.2. CHEMICAL SEPARATIONS

This section discusses the technologies available to perform solvent extraction operations within the reprocessing plant. Centrifugal contactors have been selected as the solvent extraction technology of choice due the enhanced separation efficiency they provide for their small plant footprint.

5.2.1. SOLVENT EXTRACTION

Solvent extraction is the key technique at the heart of all commercial aqueous reprocessing technologies. It relies on the selective transfer (via preferential dissolution) of species from an aqueous stream to an organic stream which can be contacted with a clean aqueous phase (under different process conditions) to transfer the extracted species back into the aqueous phase. Solvent extraction is also utilised in the solvent recycle streams as well as in some waste treatment processes to remove specific species such as silver.

There are three main types of solvent extraction equipment used in the nuclear industry: Mixer settlers, pulsed columns, and centrifugal contactors. Each are designed to provide efficient mass transfer by creating fine dispersions of droplets of one phase (the dispersed phase) in the other (continuous phase) which increases the surface area available for mass transfer. Separation of the phases is then performed, utilising the difference in phase densities either via gravity (a time dependent process) or by applying a force to the mixture e.g. centrifugal force. The size of droplet formed in the mixing phase also has a significant effect on the rate of phase separation, with finer droplets taking longer to coalesce and separate than larger ones.

All techniques discussed can in some way be configured to allow a stage-wise extraction (where the product of one separation stage becomes the feed for the next, improving extraction efficiencies). This is necessary as the extraction kinetics are usually slow, to the point where more than one stage is required to reach the desired extraction.

5.2.1.1. MIXER-SETTLERS

The mixer-settler is one of the simplest contactor designs available. They have been adopted in early reprocessing plants such as the Windscale and Magnox reprocessing plants at Sellafield, UK. This equipment comprises of a mixing vessel where two phases (aqueous and organic) are contacted in a continuous manner (an impeller provides agitation and a pumping effect), and a settling tank (Figure 18). The dispersion from the mixer passes via a weir into the adjoined settling tank, sized appropriately so that the two phases separate via gravity, before flowing out of separate weirs.

Mixer settlers are an extremely mature technology and can be built into banks of significant size to improve extraction efficiency. However, the design involves mechanical drives which can fail and cause maintenance issues. In addition, it can be difficult to design the equipment to criticality safe geometries therefore some sort of neutron poison may have to be added to the liquor. Whilst this technology has performed well over time, higher efficiencies (% recovery for a given footprint) can be obtained using the other types of contactor discussed.

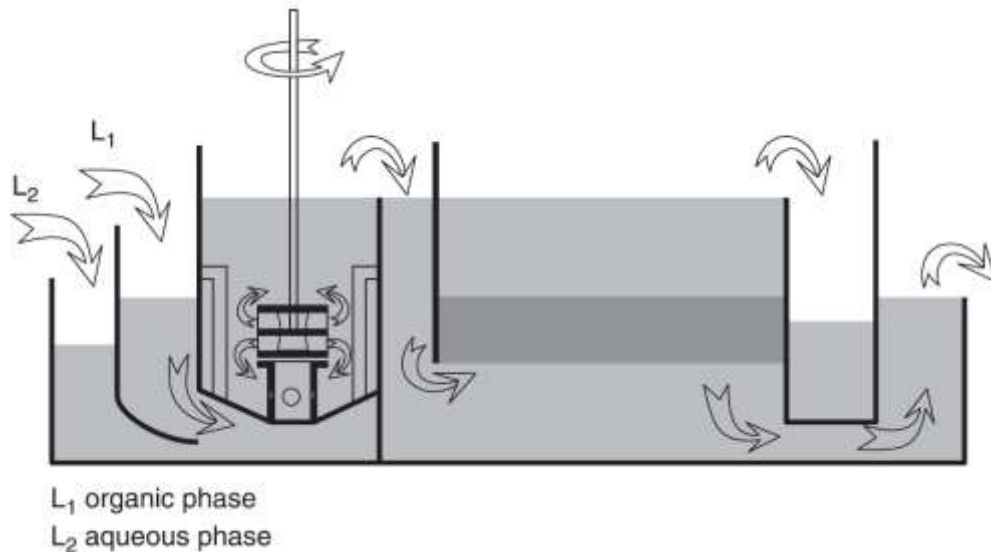


Figure 18 - Schematic of a single mixer-settler unit [23]

5.2.1.2. PULSED COLUMNS

THORP utilises pulsed columns in its primary separation solvent extraction cycle. These were originally adopted due to additional design constraints introduced by the higher fissile content of the fuel relative to earlier reprocessing plants which posed a criticality hazard. In addition, the combination of higher activity and the long residence times within the settling section of mixer settlers would have resulted in high levels of solvent degradation.

Pulsed columns consist of narrow, tall columns with a number of perforated plates positioned at equal intervals along its height. The two phases are added at the top and bottom of the column, with the less dense liquid being introduced at the bottom. The two phases move to the opposite end of the column via gravity effects. The space between the perforated plates simulates the separation stages like that of a mixer settler, with the two phases being contacted counter currently in each stage. A pneumatic limb provides an upward pulsing effect which forces the lighter liquid through the holes of the plates into the heavier phase of the stage above. The phases then separate via gravity before the pulsing repeats. The size of the holes determines the droplet size whereas the distance between plates determines the size of the settling “tank”. The geometry of the column is such that a criticality cannot happen under any circumstance.

Pulsed columns require a much smaller plant footprint compared to mixer settlers whilst also offering increased extraction efficiency (% recovery) with no moving parts. However, these units can be very tall often occupying several stories of plant space.

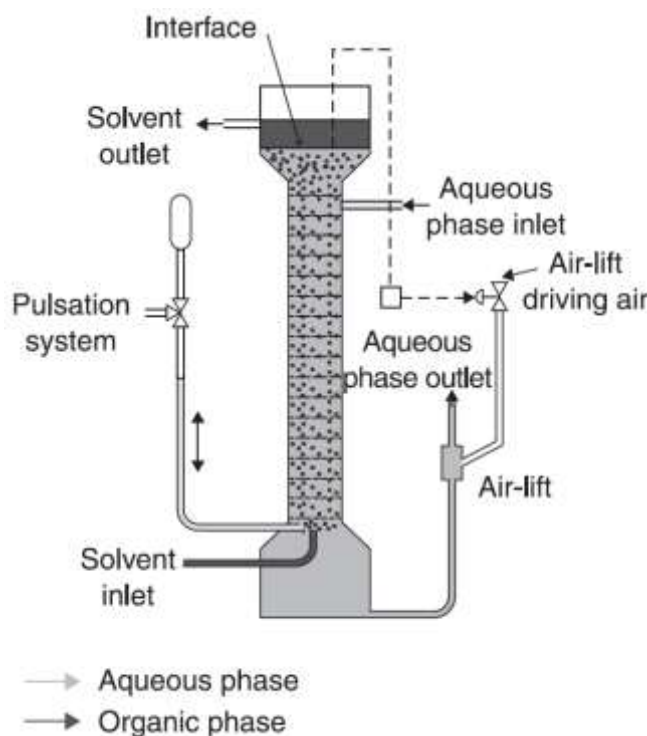


Figure 19 - Schematic of a pulsed column [23]

5.2.1.3. CENTRIFUGAL CONTACTORS

Centrifugal contactors offer enhanced extraction due to the use of centripetal force instead of gravity, significantly increasing the rate of phase separation. Relative to the other types of solvent extraction equipment, centrifugal contactors are very compact in size and can occupy significantly less plant footprint for a given level of extraction, offering a reduction in overall plant footprint and costs. The principle remains the same as that for other types of contactor – the two phases are agitated so that a dispersion of droplets is formed, the resulting dispersion is then spun at high speed so that the heavier phase travels further towards the periphery and the two phases can be taken off separately. A rotor typically provides the liquid with rotational momentum in the outer annulus. The mixture is then drawn into the bottom of the rotor via static radial vanes. These provide a dispersion of droplets which then separate back into heavy and light phases within the rotor.

There are two main types of centrifugal contactor: staged and differential. Staged contactors involve the introduction and mixing of both phases at the periphery, the two phases then travel through the contactor and are separated. To achieve a counter-current flow arrangement the contactors must be arranged in a stage-wise fashion. Differential contactors contact the two phases counter-currently within a single contactor. The lighter phase is introduced at the periphery and the heavy phase at the centre. As the liquid spins the two phases are contacted and separated.

The control of the hydrodynamics is vital to efficient operation of the contactors. Factors which will affect this are the pressure inside the contactor and of the feeds, the holdup within the contactor, the extent of flooding and the degree of back-mixing.

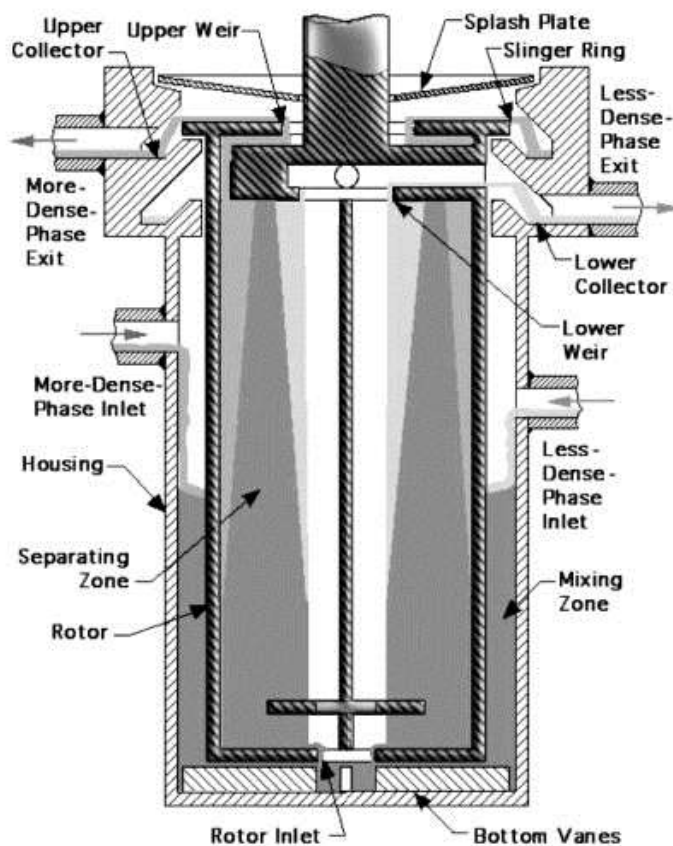


Figure 20 - Mixer settler cross-section [28]



Figure 21 - Mixer settler in a nuclear installation [24]

5.2.1.4. SILVER RECOVERY

Enhanced dissolution techniques will give rise to aqueous silver nitrate. This will be routed with aqueous fission products in the solvent extraction cycles and leave the system with the aqueous fission product stream. The silver can be recovered from this stream and recycled, reducing operational costs, HLW volumes and the risk of insoluble forming through reactions with any halides present.

To recover the silver the aqueous fission product waste stream must be contacted with odourless kerosene to remove any solvent entrained with the stream. An electroplating unit has been suggested in previous designs as a method for extracting the silver as a metal, with the aqueous raffinate proceeding to HLW treatment. This technology will be employed in this design. The unit can be washed with nitric acid to remove any residual solids before the silver is re-dissolved in 4.5M nitric acid at elevated temperatures to provide a recycle feed to an enhanced dissolver. This process must be performed in batches therefore multiple units may be required to satisfy throughput.

5.2.1.5. SOLVENT RECYCLING

The recycling of solvent within the solvent extraction cycles is expected to be very similar to that already adopted within commercial reprocessing plants, the key difference again being design for the higher concentrations of fissile material within the system. The higher levels of radiation are expected to increase the rate of solvent degradation. The system must therefore enable for the monitoring and purging of degraded solvent to prevent a build-up of degradation product.

Solvent will be contacted with alkaline carbonate and weak acid solutions. The alkaline carbonate removes residual acids entrained in the solvent whilst the acid wash in turn removes any entrained alkaline carbonate. The solution is then filtered and stored prior to reuse, to allow for monitoring and purging. A fresh top-up solvent feed would be introduced into the holding tank to ensure good mixing.

5.3. FINISHING

5.3.1. URANIUM FINISHING

Uranium finishing techniques are well established and as such it is assumed that U finishing techniques utilised in THORP (thermal denitration) will be suitable for fast reactor fuel.

In THORP the U-rich aqueous stream from chemical separation flows to a safe-by-shape buffer tank which allows for monitoring, sampling and blending with low enriched uranium (from an external source). The blending of uranium is crucial so that the concentration of fissile ^{235}U will be sufficiently low post concentration via evaporation to avoid the risk of criticality. The criticality safety philosophy changes from safe-by-shape to safe-by-mass at the evaporation stage. The solution flows through a steam strip column prior to feeding the evaporator.

Blended uranyl nitrate feeds the evaporation stage which concentrates the stream from to approximately 1000gU/L. The crystallisation temperature of the concentrate is approximately 60°C therefore the remainder of this processing line is trace heated to a minimum of approximately 90°C. The evaporator vapour flows into the steam stripper to remove any solvent that could combust and is then condensed and sent to low active (LA) effluent treatment.

Concentrate is stored in buffer tanks prior to thermal denitration in a fluidised bed reactor. At this point sulphuric acid is added to improve flow properties for downstream operations. Within the fluidised bed reactor uranyl nitrate is thermally decomposed to UO_3 powder using heated air. The UO_3 then overflows into a second fluidised bed “pre-cooler” where it is cooled by air to approximately 150 °C. The powder is conveyed to a hopper by air at ambient temperature which further cools the powder to around 50 °C.

Powder product is screw-fed into drums, lidded and sealed. It then undergoes monitoring for contamination and enrichment level.

5.3.2. MIXED ACTINIDE FINISHING

The actinide finishing line is expected to resemble that of the Pu finishing line in THORP however some key alterations have been identified in previous GANEX concept designs, these being the addition of an organic

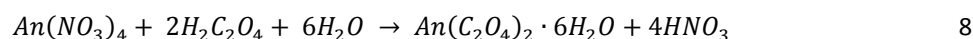
destruction stage and a valence conditioning stage [4] [2]. It should be noted however that the detailed understanding of this process is significantly less developed than other sections of the overall process.

The core process will comprise of organic destruction, valence conditioning, evaporation, precipitation (with oxalic acid), filtration and furnace/calcination processes. The aqueous mixed actinide product stream from chemical separation can contain Acetyl hydroxamic acid (AHA) and Bis tryazinyl pyridine (BTP) molecules (used to selectively strip out the actinides), which may create “red-oil” safety concerns in the evaporator. A Red Oil Excursion (ROE) is an explosive, runaway nitration-oxidation reaction [29]. In addition the presence of organic can cause issues within the precipitator when the actinides are separated from the oxalic acid as the actinides-organic bond is much stronger than the actinide-oxalic acid bond. Therefore the stream must undergo organic destruction prior to traditional oxalate precipitation finishing. A number of techniques have been identified for the destruction of organics however this area requires further research before a detailed design can be produced. Some technologies identified include an electrochemical processing or ozone sparging.

After organic destruction the liquor will require acid conditioning, to ensure the nitric acid concentration is suitable for evaporation.

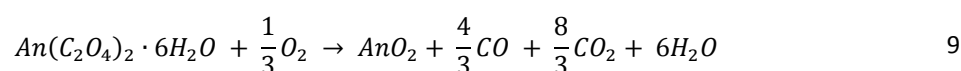
The oxidation state of the solution produced in the evaporator has a significant effect on the shape and size of the crystals formed during precipitation. The valence conditioning stage ensures that the actinide oxide powder produced has the desired uniform crystal structure, as this is crucial in the fuel manufacturing process. Similar to the organic destruction stage, more research in this area is required prior to the detailed design of a plant. It is likely that an electrochemical process may be one option, however for flowsheeting purposes a hydrogen peroxide oxidation unit is assumed.

The solution is evaporated, concentrated and buffer stored prior to precipitation. The evaporator off gas is condensed and treated as effluent waste. The precipitation of the actinides with oxalic acid will occur at temperatures approximating 60°C and will follow a similar mechanism to that of Pu (see equation 8).



The slurry formed in the precipitator overflows to the filtration stage, typically performed in a rotary vacuum filter. The liquor is removed using a low vacuum, and then the slurry is washed with dilute acid and water before being subject to a high vacuum which dries and compresses the particles into a cake. The filter cake is scraped off and transported via a screw feed to furnace processes.

The actinide-oxalates are fed through a drying furnace by two intermeshing helical screws. An air and argon mixture contacts the material in a counter-current fashion drying and converting the oxalates to dioxide powder form (see equation 9). The temperature of the furnace varies from ~100 °C at the powder inlet to ~400 °C at the gas inlet.



The product powder from the drying furnace feeds the calcination furnace. This has a much larger residence time compared to the drying section of the furnace to ensure the product meets the required moisture content and to remove any remaining volatile components. The powder is again moved through the furnace by a rotating screw.

The powder is blended to ensure a homogenous mixture of isotopes before being loaded into screw-top product cans. These cans are placed into an additional two layers of overpack, welded and marked with barcode identification. The cans are then monitored and sent to storage.

6. WASTE AND EFFLUENT

6.1. SOLID WASTE

The plant will generate solid low level waste (LLW), plutonium contaminated material (PCM), and intermediate level waste (ILW).

LLW is anticipated to originate predominantly from operation of the plant. This will be collected in waste containers and exported to an on-site waste treatment facility for compaction and repackaging. The waste will then most likely be stored in a low level waste repository.

PCM will be buffer stored and exported to a dedicated on-site engineered storage facility. Failed equipment from in-cell will be decontaminated and disposed of as PCM.

Hulls from head end dismantling and shearing are expected to constitute solid ILW. These will be packaged, buffer stored and exported to a dedicated on-site waste treatment plant for encapsulation into a cement matrix waste-form. Wetted centrifuge cake containing insoluble fission products is also anticipated to be disposed of as ILW, however it is possible that this could be classed as HLW depending on its composition. Further research is needed to assess which waste category the cake will fall into.

6.2. LIQUID EFFLUENT

Liquid effluents are generated in the chemical separation and finishing sections of plant. These will be categorised into high level waste (HLW) and low active (LA) effluent. Degraded solvent, OML, acids, alkali and lanthanide effluent will all require further processing in separate facilities.

High level waste streams will contain fission products and will be exported from the facility via pipe-bridge to be dealt with directly by an adjoining waste treatment facility. The HLW will be concentrated and decay stored prior to being immobilised, most likely through vitrification in glass. This product will be interim stored prior to long term storage in a Geological Disposal Facility (GDF). Low Active (LA) effluent, which includes acidic and alkaline raffinate will be buffer stored and sent to an appropriate waste treatment plant on the same site. Degraded solvent is sent to an on-site solvent treatment facility.

6.3. GASEOUS EFFLUENT

Gaseous effluent will be generated in the head end and finishing sections of the plant.

A centralised off gas system will be utilised for the continuous, backup and silver dissolvers whereas a separate off gas system will be used for the voloxidation off gas. Further off gas treatment units will be required for the TDN and calcine gaseous effluent. All of the off gas systems will eventually feed into a common filter bank and stack.

The gaseous discharge is expected to conform to the required regulation of the site however further investigation into this is required at further design stages as the exact limits are not known at this time.

7. FLOWSHEETING

This section should be read in conjunction with the output data illustrated in Appendix 11.1

The flowsheet for this plant was created using gPROMS process modelling software and, where possible, was built upon existing Euro-GANEX models developed through the SACSESS project [2]. The latest Euro-GANEX model, was of an engineering/pilot plant scale and utilised hot-test data obtained through NNL research [30]. The previous model however, only considered the chemical separation section of plant. The model developed for this concept design not only considers head end and finishing sections of the plant but does so at the industrial scale.

The model tracks the mass of species throughout the process and outputs stream data such as mass flow, molar flow, mass concentration, molar concentration, total mass flow, density and total volume flow. The presence of solids within the head-end section of plant has complicated the prediction of stream density, as a result, there is less confidence associated with the volume flows of these streams however the results appear reasonable. The requirement for further development with the density calculations has been carried forward as a recommendation (see section 9).

Appendix 11.1 holds summary tables of the pertinent flowsheet output data. The summary tables illustrate the mass of elemental metals (not the metal compounds), and other reagents.

The new model includes a slightly updated component list. From the FISPIN data, elements were grouped to form the species list shown in Table 4. Compared to the old models, this version now considers curium but does not consider ^{14}C due to lack of data.

The model produces a flowsheet that is representative of the flows expected throughout the process plant. It does, where possible, represent every major process unit, but not all of them. The model also assumes a completely continuous process and doesn't take account of residence times in buffer storage tanks or semi-batch operation. The model is of the steady state variety - meaning that the flowsheet produced is representative of a snapshot in time during continuous operation. It does not consider start up, shutdown or maloperations.

The Chemical separation section of the flowsheet has been based on lab-scale hot test data produced in a previous project [30]. It is evident from the summary tables that the values used do not scale well as there appears to be a lot of excess water and in some cases solvent (where the quantity of solvent is dependent on the aqueous flow). Therefore the flowrates in this section are higher than anticipated. Efforts could be made to increase the concentrations of these streams however further experimentation would need to be performed to support this.

The concentration of U in the chemical separation feed has been adjusted to align with the hot test data (approximately 100g/l). In reality this stream would be as concentrated as possible to match the rate at which the solvent can be loaded with heavy metal. This concentration allows for a direct comparison with the decontamination factors derived from the hot test data and used in the SACSESS flowsheeting exercise.

The flowsheet utilises the decontamination factors from the SACSESS work to determine the routing of species through the plant. In addition, volumetric ratios were determined and used in conjunction with heavy metal mass flowrates to determine solvent and aqueous flowrates. Upon review of the summary data, this strategy has produced a flowsheet which would, in theory, function. It is however in need of further optimisation which

is only possibly through iterative phases of work. The dilute nature of the streams is believed to be the result of solvent to aqueous ratios not scaling well. More information is required regarding how the solvent to aqueous ratios will scale up, and the mechanism by which the solvent can be loaded to its maximum to enable the streams to concentrate the streams.

Future work should consider the use of a model which calculates the mass transfer based on distribution factors (which determine how a species interacts with each phase) and mass transfer principles. However, these would require a better understanding of the scale up of the hydrodynamics within centrifugal contactors.

The high flow rates in chemical separation are carried through to the evaporation stages of the finishing lines however all excess water is removed here, leaving representative flows of material throughout finishing. Equally, the high flow rate of aqueous fission product should not impact on the volumes of HLW generated, as the excess liquor would be removed in an evaporation stage through HLW treatment and storage.

The output of this flowsheet does not reflect what would be carried forward to the final design of a plant. Instead, it provides indicative flows of species underpinned by existing hot test data, which should act as a first iteration of a full scale flowsheet, and identifies potential gaps in knowledge which, if filled, would progress design capability in this area.

7.1. FUEL FEED

As discussed previously, the feed to the plant is assumed to be spent fast reactor MOX. The 331 pin ASTRID concept fuel was used as a basis for this design.

The fuel input to the model assumed a mock-fuel comprising of a mixture of breeder and driver fuel. The exact composition of this fuel was calculated based on two FISPIN model outputs. This better represents reality as one would expect either the fuel pins to contain an axial blanket of breeder fuel and/or fuel pins from the radial blanket to be reprocessed with those from the centre of the core. Table 3 shows the key parameters of the fuel used.

Table 3 - Mock Fuel Data

	Driver	Breeder	Combined (60%w/w Driver, 40%w/w Breeder)
Cooling Period /years	4	4	4
Burnup /GWd/tHM	117	23	79.4

To simplify the tracking of species throughout the model, species which behave similarly were grouped as follows (all other species are modelled separately):

- Volatile Fission Products (VFPs)
 - Kr, Xe
- Semi-Volatile Fission Products (SVFPs)
 - Tc, Ru, Te, Mo, Rh
- Fission products in various oxidation states (M, MO, MO₂)
 - (MO): Sr, Ba, Ra
 - (MO₂): Zr
 - (M): Ag, Pd, (misc of minute mass)
- Lanthanides in the M₂O₃ form

- La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu

For each group, weighted molecular weights were used (based on the quantity of each isotope present). Corrected mass fractions were calculated based on the mass of oxides and cladding present, which the model takes as input parameters.

For simplicity, any species that makes up less than 0.1% of the mass of fuel (from the FISPIN output data) was ignored. This introduces a small but negligible error in the calculated mass fractions.

Table 4 - Model Input Data Summary (Grouped species in orange)

	Mass of comp (kg)	Mass Fraction	Corrected mass fraction due to Oxygen	Altered Mass Fraction due to clad	weighted Molecular Weight (kmol/kg)
U	7.77E+02	7.77E-01	7.77E-01	2.08E-01	2.38E+02
Pu	1.36E+02	1.36E-01	1.36E-01	3.64E-02	2.39E+02
Np	3.31E-01	3.31E-04	3.31E-04	8.89E-05	2.37E+02
Am	4.95E+00	4.95E-03	4.93E-03	1.32E-03	2.42E+02
Cm	5.16E-01	5.16E-04	4.99E-04	1.34E-04	2.44E+02
Cs	8.68E+00	8.68E-03	8.10E-03	2.17E-03	1.35E+02
I	6.82E-01	6.82E-04	6.01E-04	1.61E-04	1.28E+02
H3	9.68E-05	9.68E-08	8.53E-08	2.29E-08	3.00E+00
VFPs	1.11E+01	1.11E-02	9.77E-03	2.62E-03	1.31E+02
SVFPs	1.95E+01	1.95E-02	2.15E-02	5.78E-03	1.03E+02
FPS(MO)	4.59E+00	4.59E-03	4.57E-03	1.23E-03	1.27E+02
FPS(MO2)	6.14E+00	6.14E-03	7.27E-03	1.95E-03	9.35E+01
FPS(M)	9.38E+00	9.38E-03	8.26E-03	2.22E-03	1.06E+02
Ln (M2O3)	2.11E+01	2.11E-02	2.16E-02	5.81E-03	1.44E+02
Clad	-	-	-	0.73	56.00
Total	1.00E+03	1	1	1	
Mass of SS clad /kg	2727.27				

7.2. MODEL FLOWSHEET

Figure 22 Illustrates the flowsheet developed in gPROMS. It is a representation of the plant at steady state and, as previously mentioned, does not represent every single unit that would be present in reality. Each box represents a processing unit or a small group of processing units logically linked to one-another. The process steps are illustrated in more detail in appendix 11.1 the figure below gives an overview of the entire process.

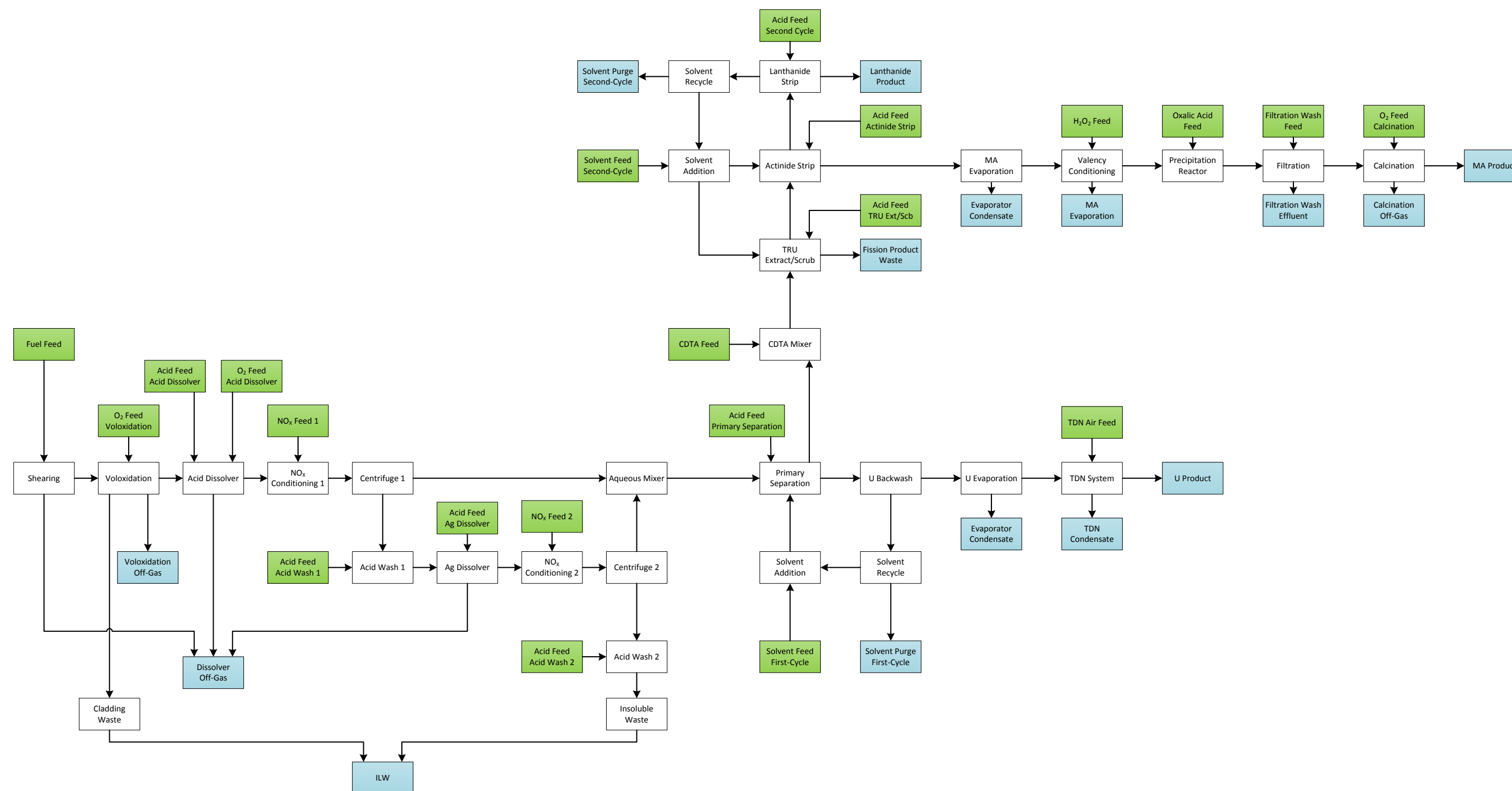


Figure 22 – Flowsheet input to gPROMS ModelBuilder software (Inputs in green, outputs in blue)

8. CONCLUSIONS

This report presents the concept design work undertaken in GENIORS Work Package 8 to produce a process description of a reprocessing plant operating a Euro-GANEX flowsheet to process spent GenIV MOX fast reactor fuel.

The plant design has been focussed in four main areas incorporating head end, chemical separation, U finishing and MA finishing. State of the art technology has been considered where appropriate alongside mature, well understood technology to enable the progression of this concept to further stages of design.

The low technical maturity of some sections of the plant, where fundamental scientific understanding is lacking, is the main barrier for engineering development. Therefore future design stages should be considered in partnership with complementary research and development projects.

Radiological shielding and environmental impact assessments were not conducted as part of this design, further understanding on the separation behaviour of all of the actinides will be required to enable these activities.

Flowsheeting operations have provided sufficient indicative data to enable initial sizing calculations for some processing units which will provide an indication of the overall scale of the plant. It is appreciated that the flows in the chemical separation section of plant may not be indicative of reality, due to limitations of the modelling calculations performed. To enable better predictions more data is required to understand how the chemical separation section will behave upon scale-up. This ideally needs to be at larger scales than existing experimental data.

9. RECOMMENDATIONS

9.1. GENERAL RECOMMENDATIONS

The development of this concept design relies on a number of assumptions which have not yet been underpinned. It is strongly advised that the following recommendations be implemented prior to the progression of this plant design.

1. A site, on which the plant will be constructed, will need to be identified prior to specific design details being finalised. These activities should consider costing and layout drawings.

A full safety assessment of the design should be carried out, including shielding and criticality assessments, prior to progressing to the preliminary design stage.

2. The exact mechanisms by which the solvent wash and recycle system, for both the first and second solvent extraction cycles, will operate are unknown. This was flowsheeted using a feed-and-bleed (purge and top up) method. Significant development is required prior to more detailed design.
3. The development of a valance conditioning unit is also required prior to detailed design. The exact mechanism by which this unit will operate was unknown and therefore it was assumed to be the same as in THORP: a H_2O_2 oxidation reaction.
4. Further work is required to understand the implications of working environments, such as cells and gloveboxes, for the specific activities outlined in this design.
5. Until the exact specification of what a commercial GenIV fast reactor MOX fuel will look like is available, the head end of the plant cannot be fully specified. This includes the conditions under which the voloxidation stage should operate. This should be taken into account in future work.
6. The design and number of centrifugal contactors per bank should be refined throughout the design process with the aid of experimental flowsheet studies.
7. Research should be undertaken to assess the compatibility of the wastes generated from this plant with existing waste treatment technologies.

9.2. FLOWSHEETING RECOMMENDATIONS

The following recommendations refer to improvements of the gPROMs model from which the flowsheet has been generated. These recommendations should improve the accuracy and representation of the plant if incorporated in future work.

1. In practice, some material from the finishing lines may be recycled back into the process e.g. OML and CML from MA Finishing and excess U may be required to alter the chemistry of the chemical separation section. These recycles were not taken into account in this iteration of the model but should be considered as an improvement to better reflect actual mass flows.
2. Upon better understanding of the solvent wash cycles, improvements to the purge and top-up models should be made.
3. Future efforts should consider enlarging the list of species currently tracked by the model. This will require scientific underpinning and hot-test data to provide decontamination factors (factors which determine the routing of species through the process units).
4. Improvements to the models to enable them to track the activities of species throughout the process should be made to allow for a robust radiological and criticality assessment.
5. The density sub-models should be upgraded to better take account of solids within the process streams; this will provide more accurate volumetric flow data.
6. The relatively large flowrates in the chemical separation section of the plant are the direct result of ratios previously developed using hot test data. These ratios have not scaled well from laboratory to industrial scale. Future work should consider the development of these ratios to higher scales to improve modelling accuracy.
7. Models which consider the distribution of species between phases, and the hydrodynamic behaviour of centrifugal contactors to calculate mass transfer should be developed to replace the current application of decontamination factors.

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11.APPENDICES

11.1. SUMMARY TABLES

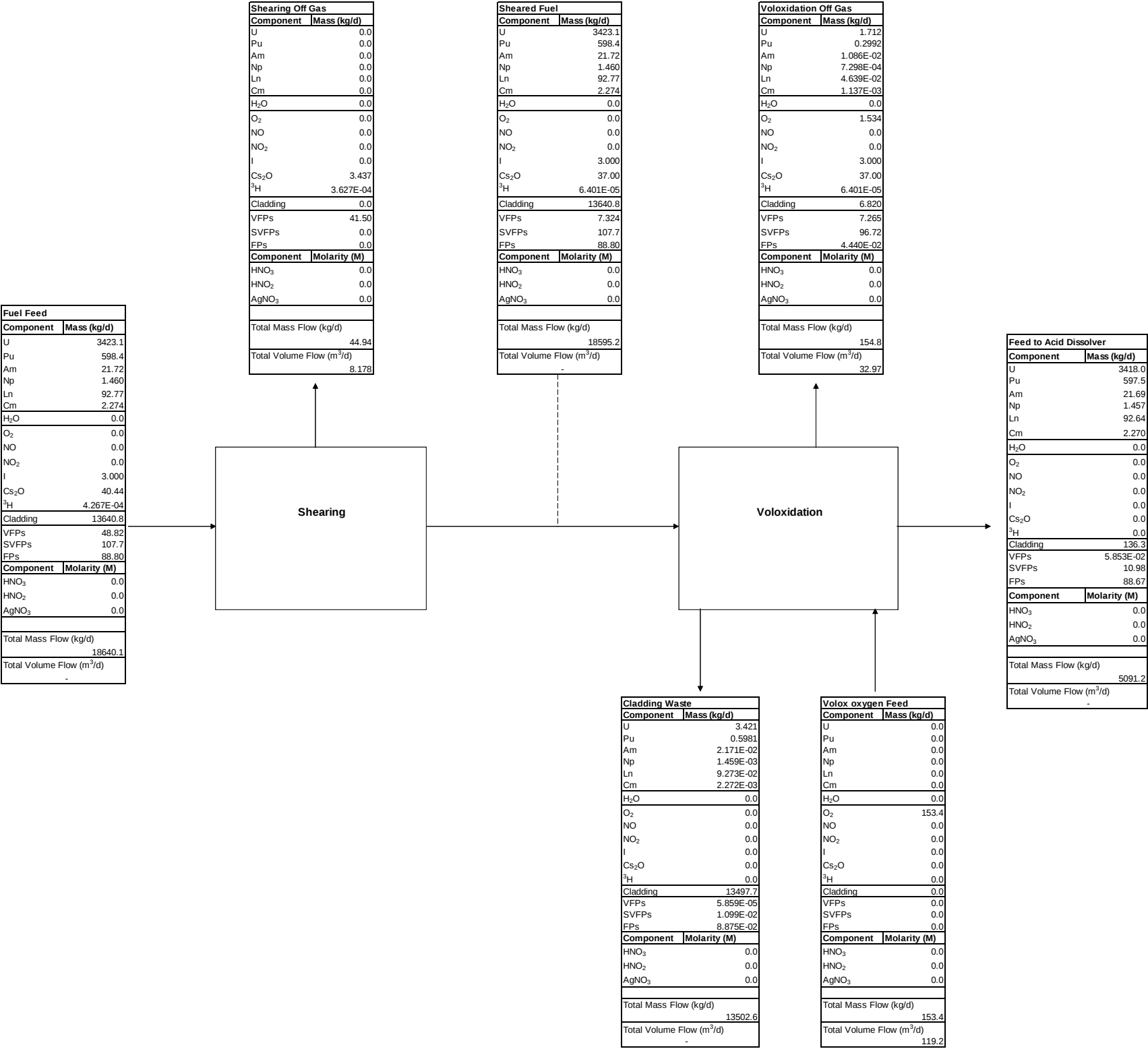


Figure 23 - Head End: Shearing and voloxidation summary tables

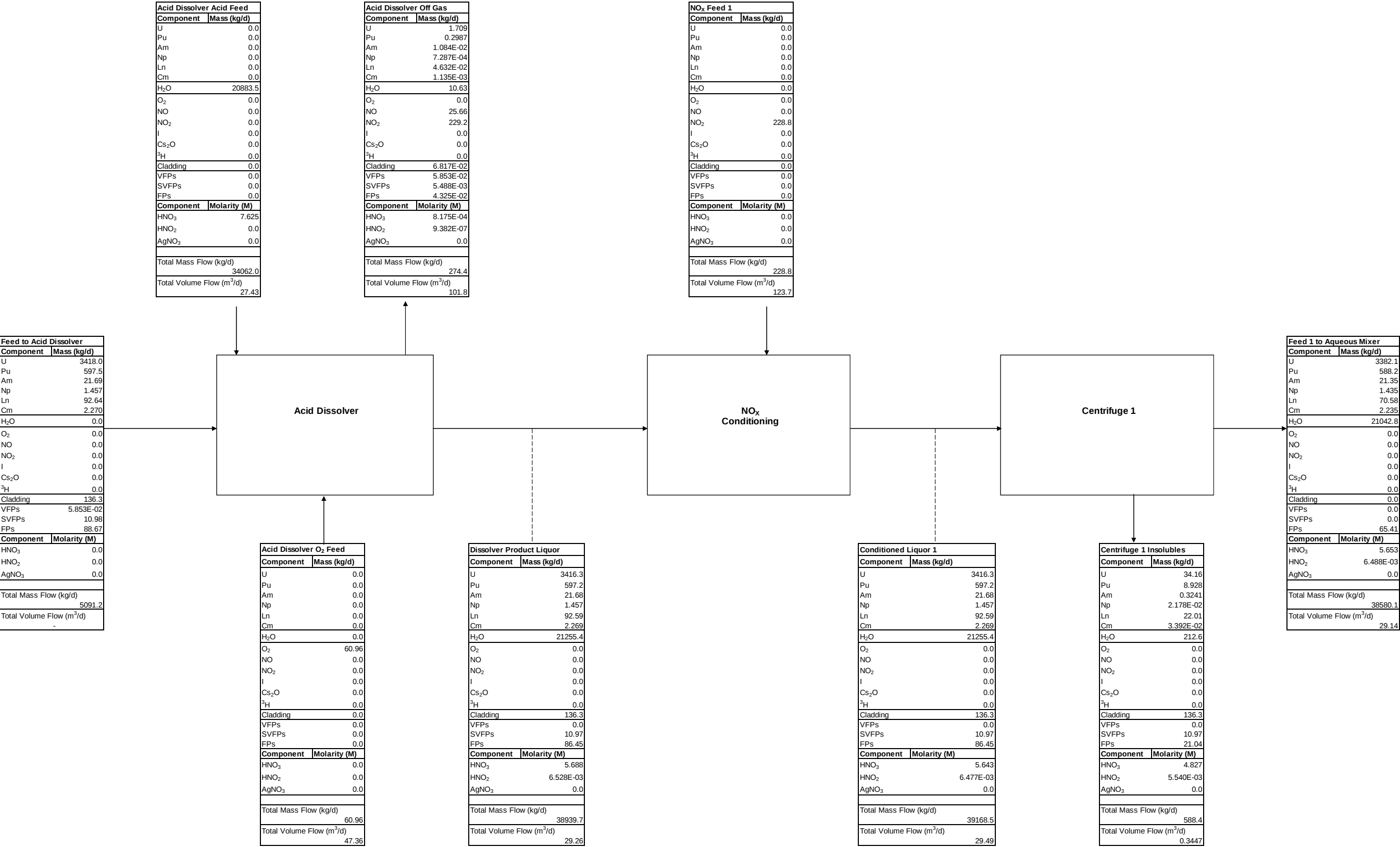


Figure 24 - Head End: Acid dissolution and clarification summary tables

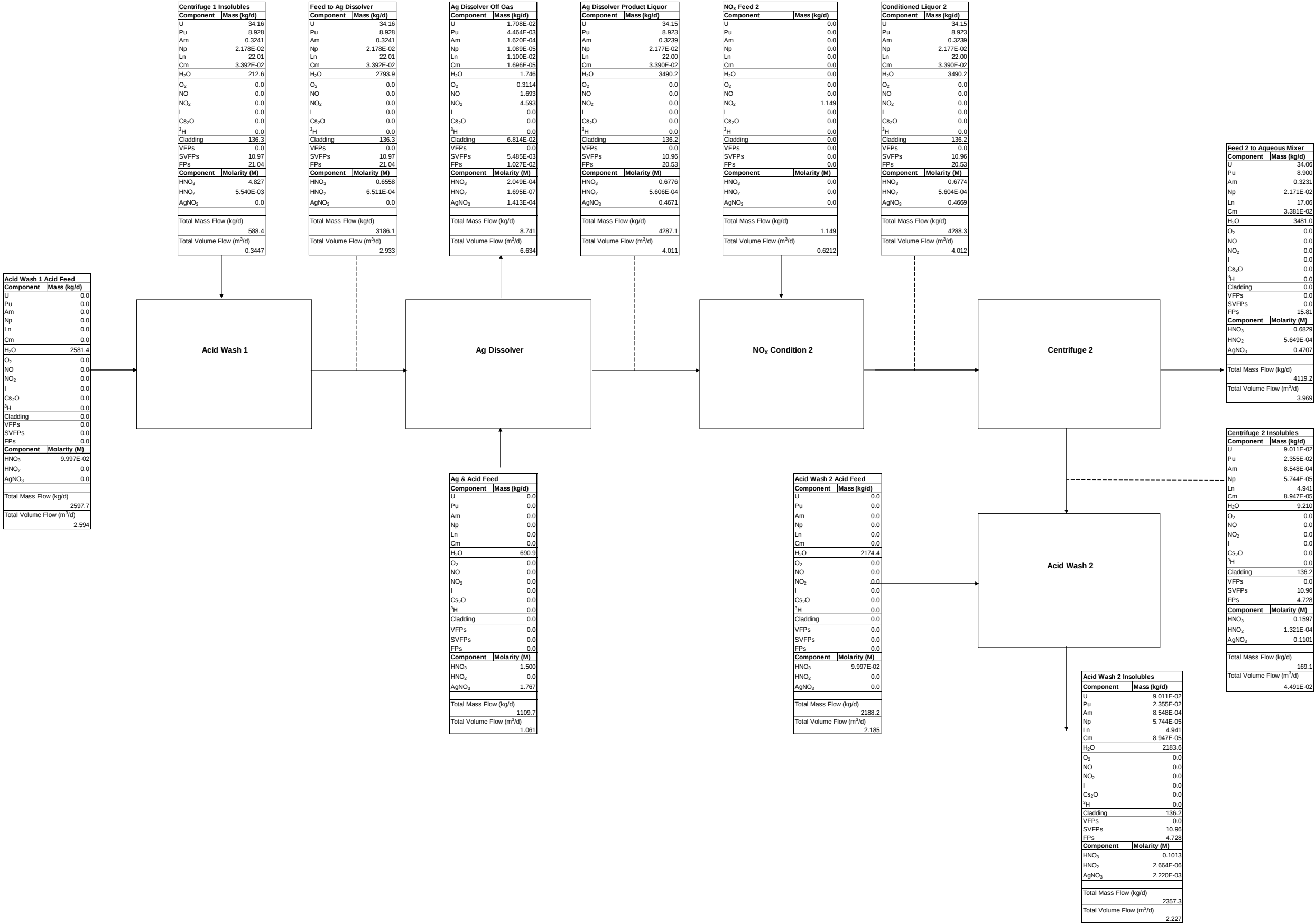


Figure 25 - Head End: Ag dissolution and clarification summary tables

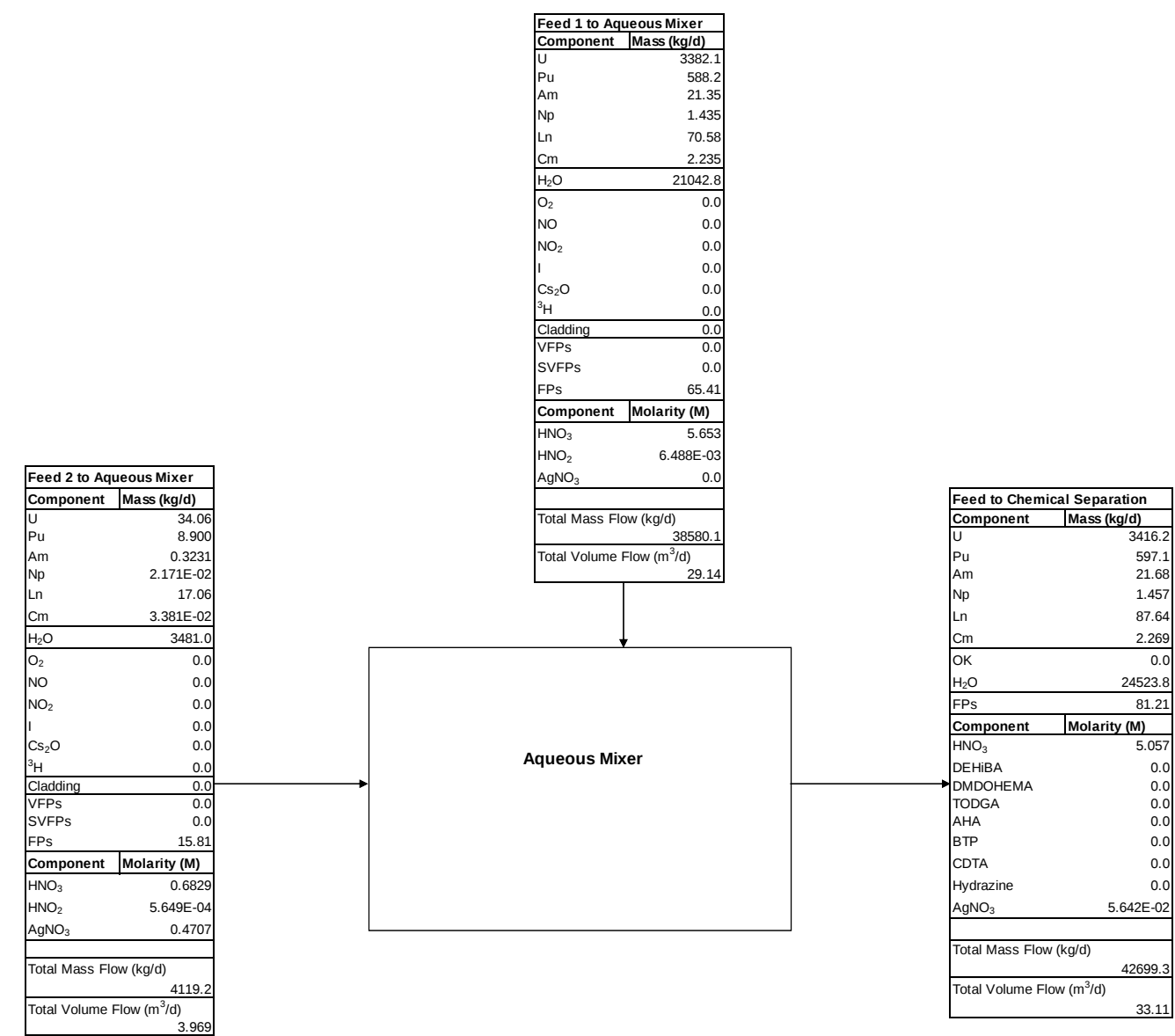


Figure 26 - Head End: Aqueous mixer summary tables

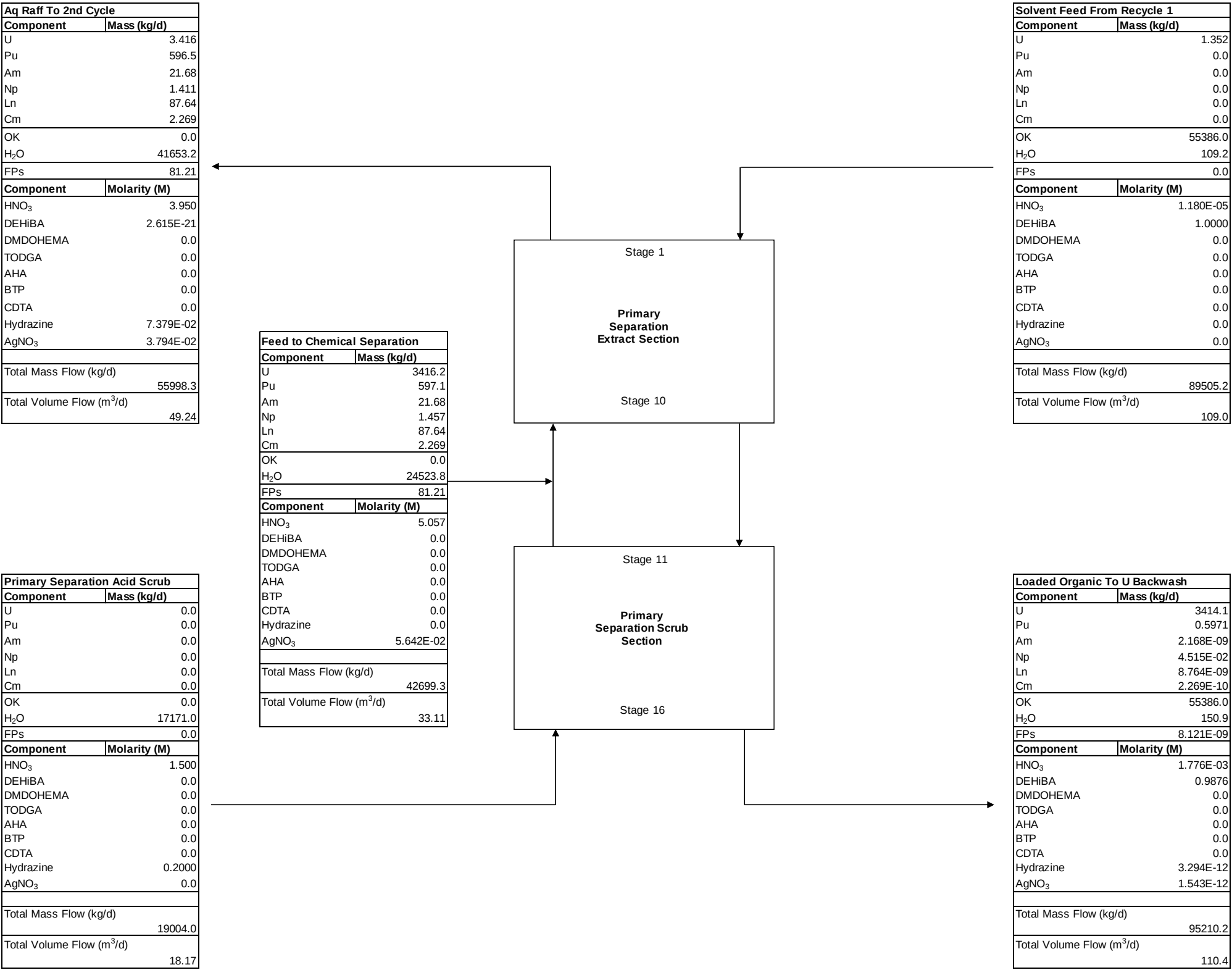


Figure 27 - Chemical Separation: Primary Separation summary tables

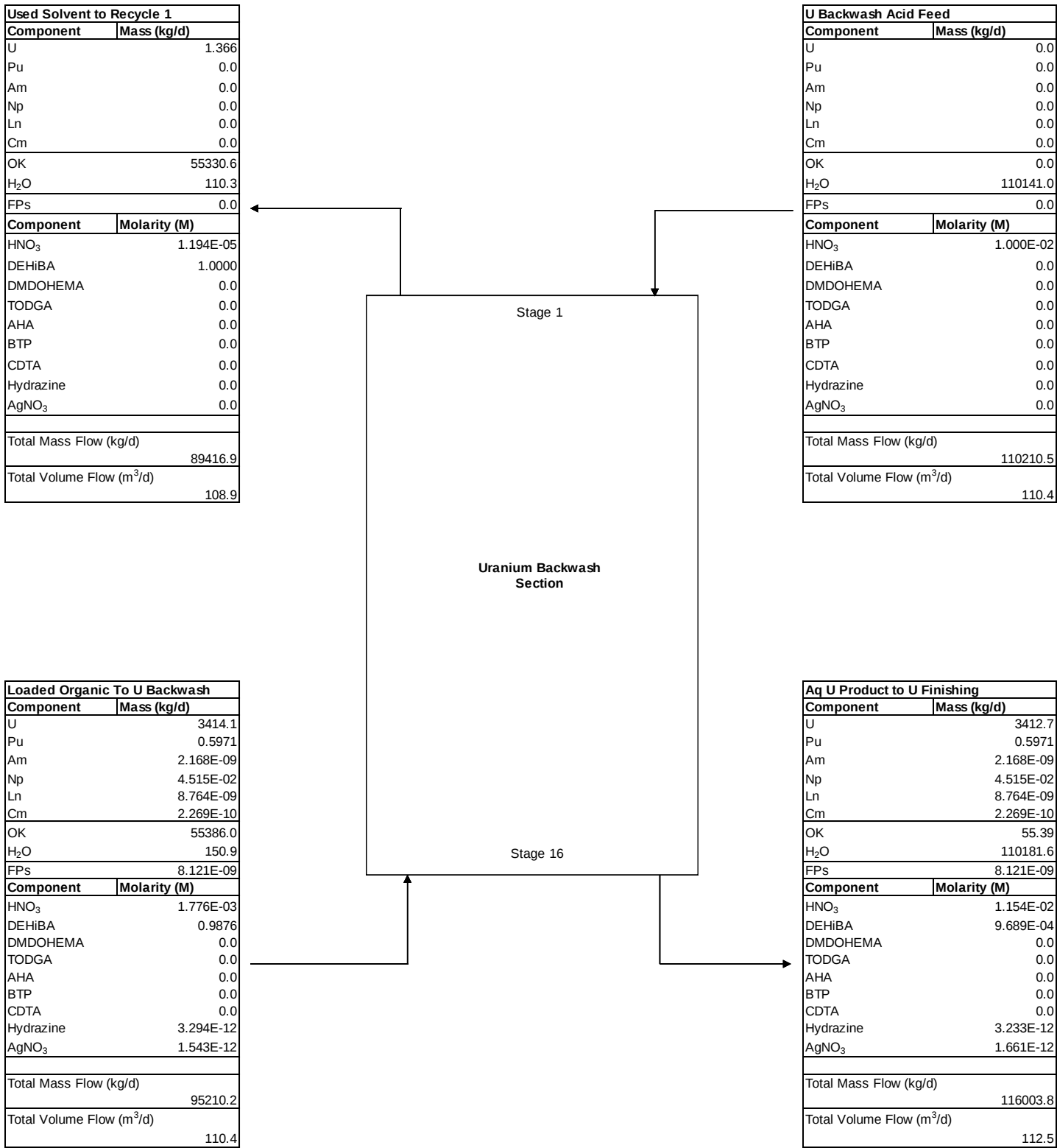


Figure 28 - Chemical Separation: U backwash summary tables

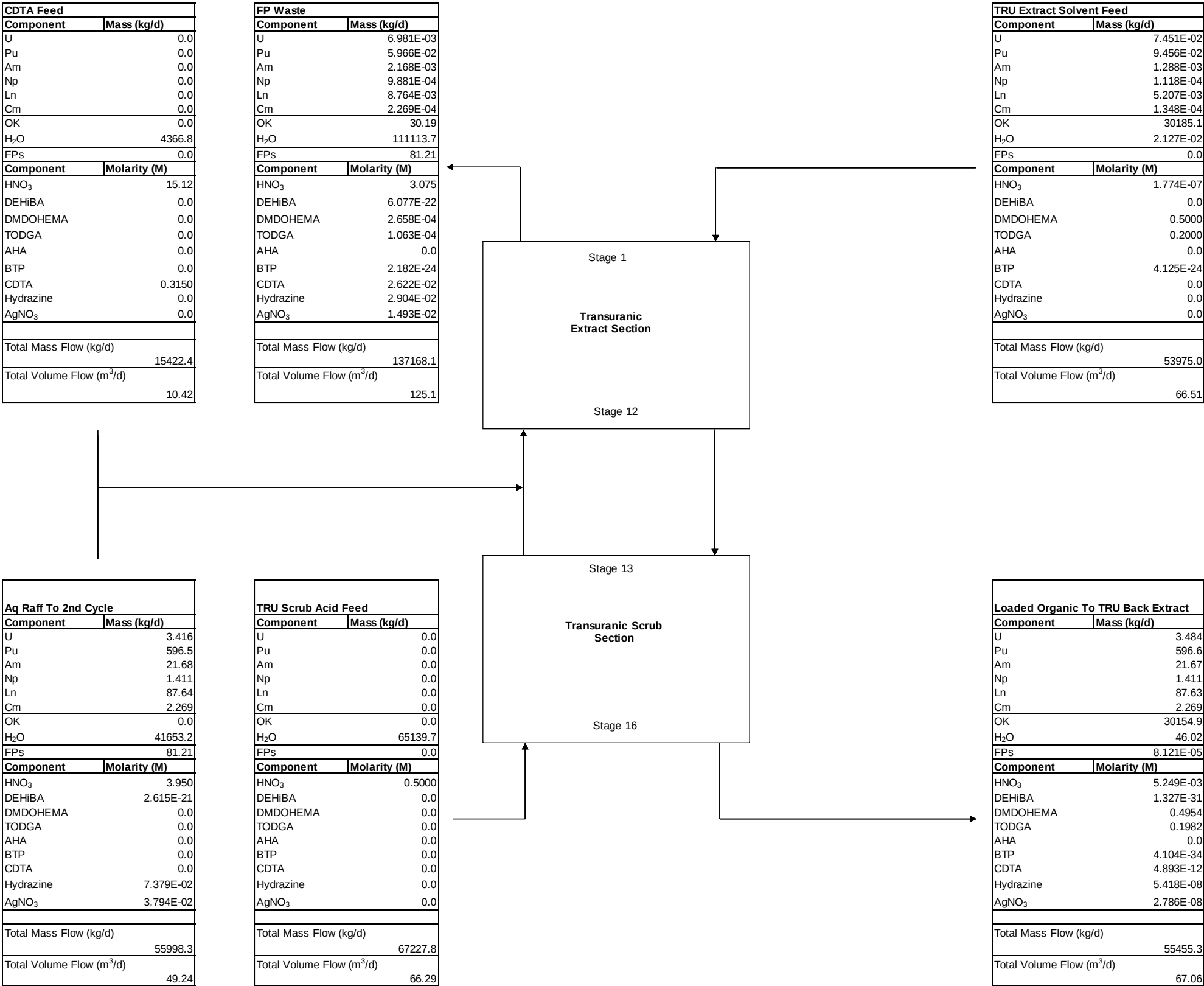


Figure 29 - Chemical Separation: Transuranic extract summary tables

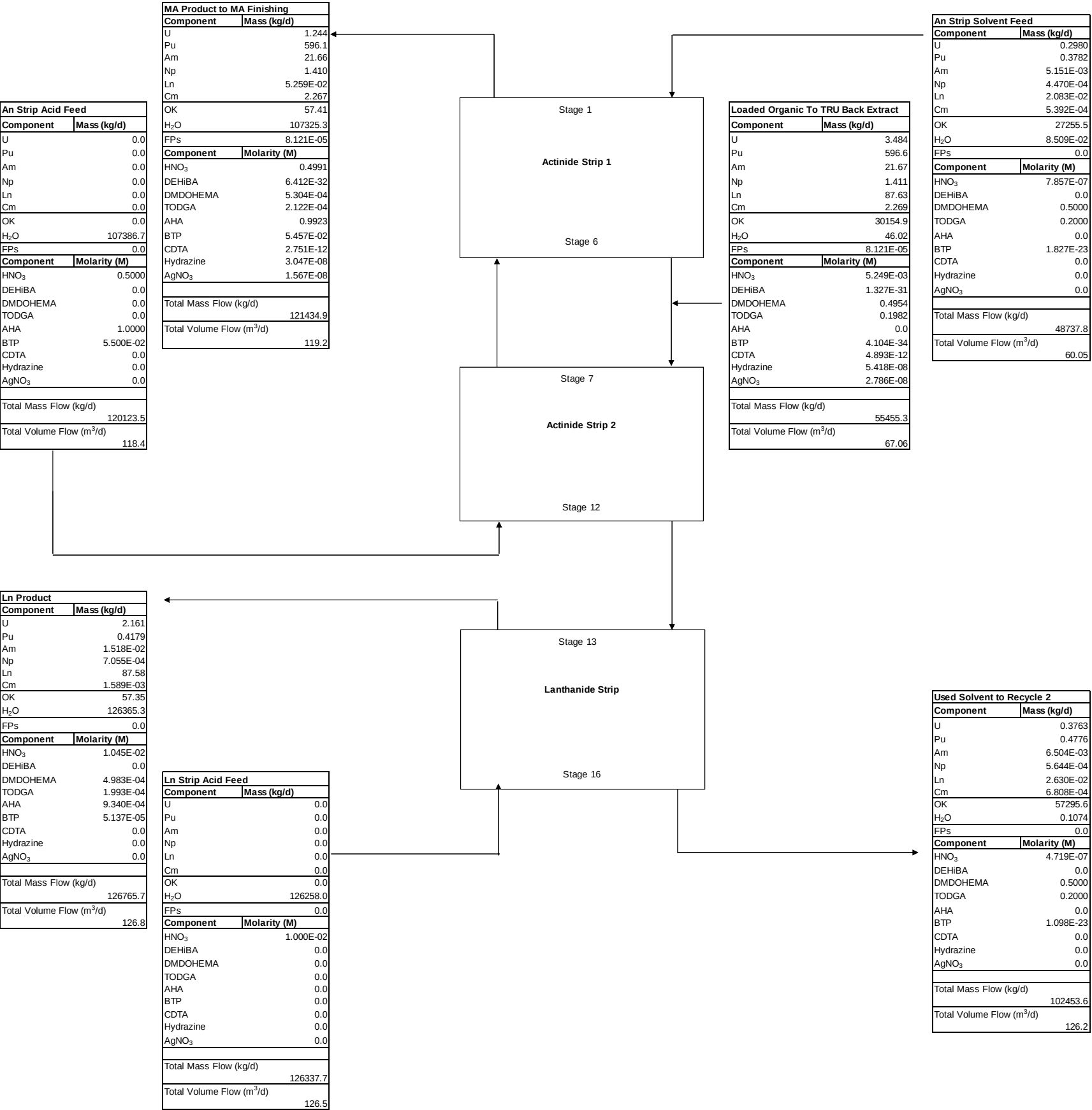


Figure 30 - Chemical Separation: Actinide strip summary tables

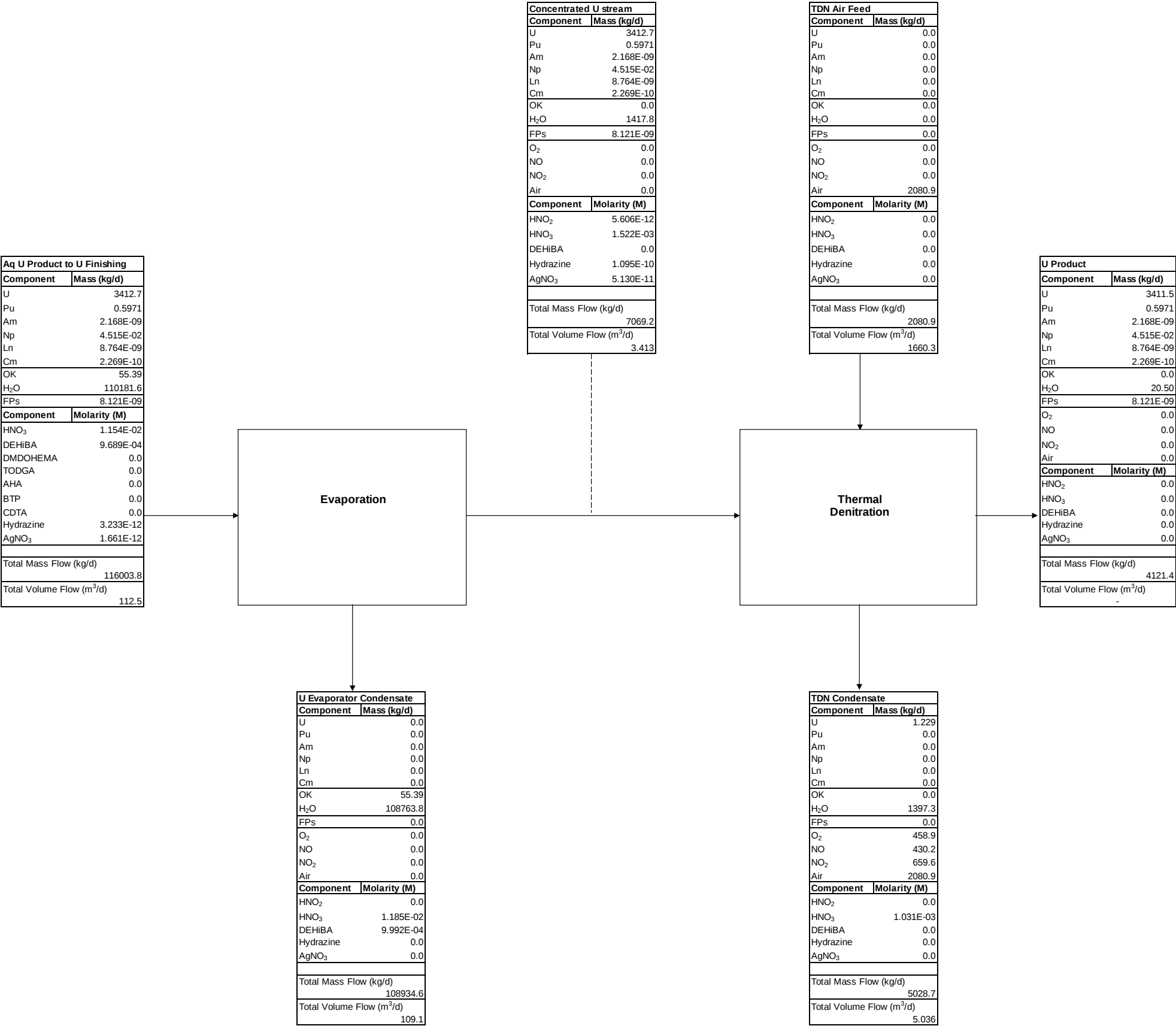


Figure 31 - U Finishing: Evaporation and thermal denitration system summary tables

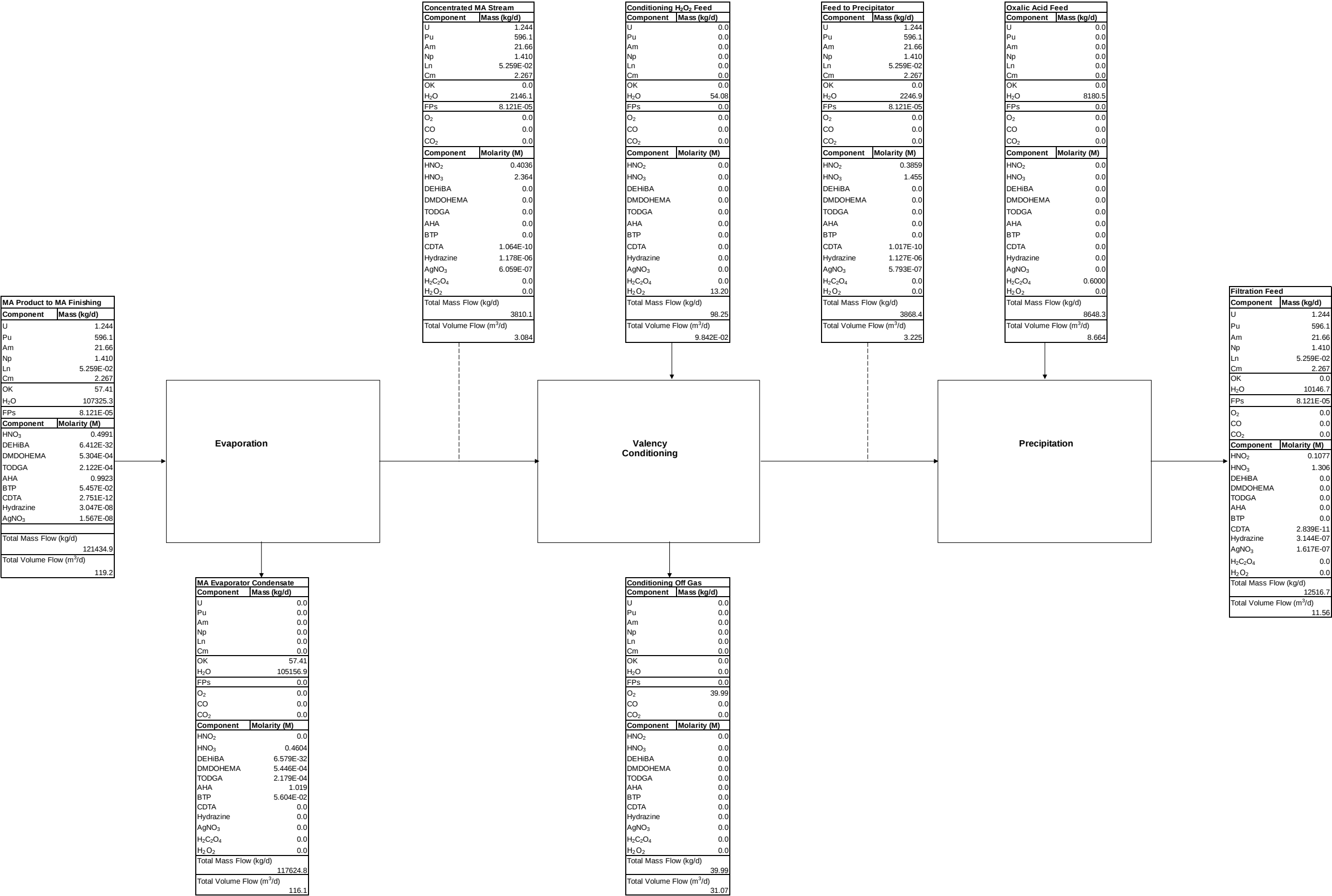


Figure 32 - Mixed Actinide Finishing: Evaporation and precipitation summary tables

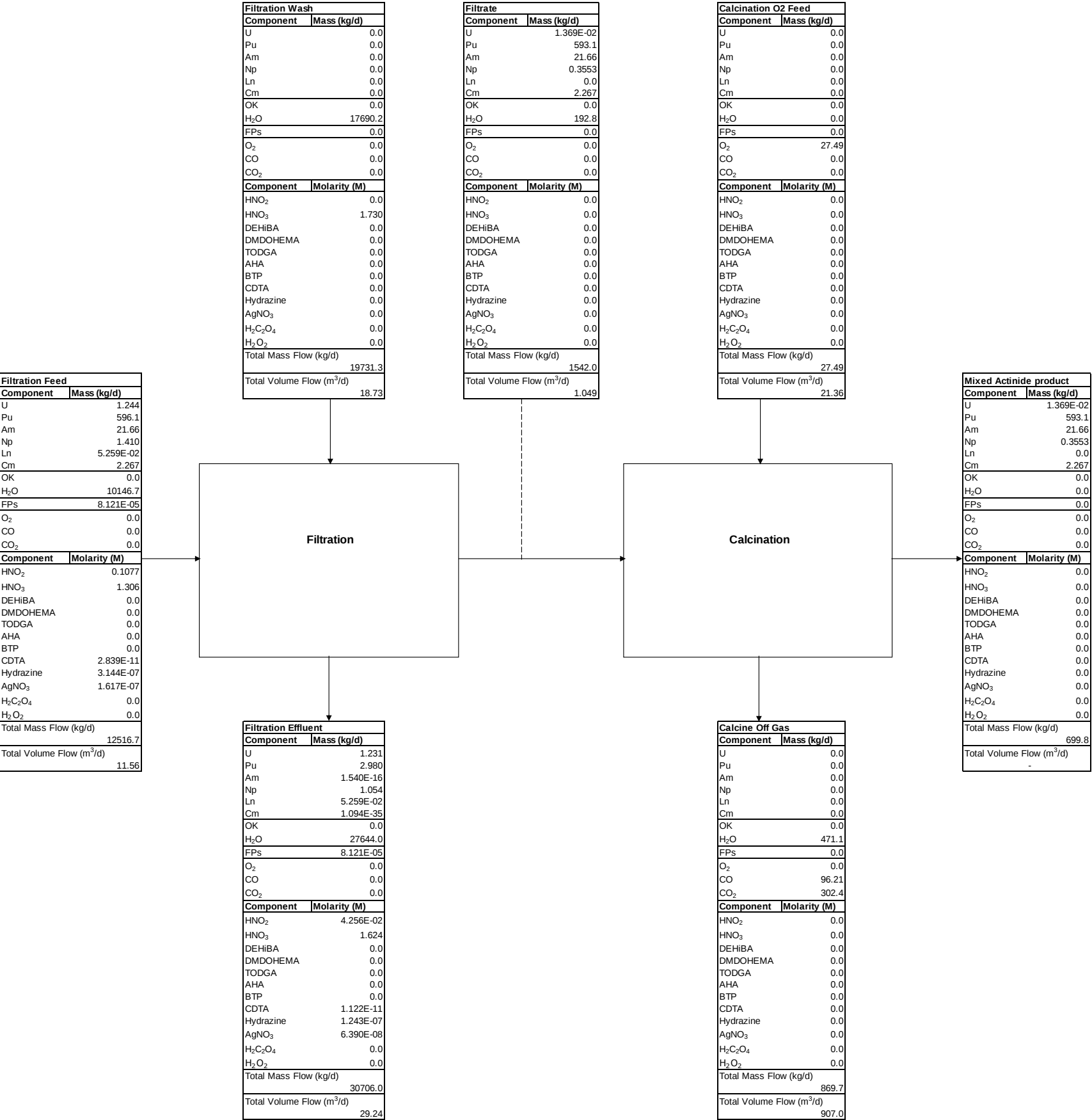


Figure 33 - Mixed Actinide Finishing: Filtration and calcination summary tables

11.2. FLOWSHEETING MODEL ASSUMPTIONS REGISTER

This Section will break down the sub-models within the model, outline each sub-model's role, and its key assumptions.

11.2.1. GENERAL ASSUMPTIONS

- The system is balanced by mass – all volumetric flows are determined by density models which calculate the density based upon the stream composition.
- Feeds to the process are “Pulled” by the unit which requires them. Excess quantities required are assigned within the model.
- Temperature and Pressure remain constant for flow sheeting purposes.

11.2.2. ACID DISSOLVER

This model represents the acid dissolution stage within the head end section of plant. Pulverised fuel from voloxidation feed this stage along with nitric acid. The outputs go to off gas and NO_x conditioning. The splits of species within this model are determined by chemical reactions.

- 100% of available U dissolves.
- 99.5% of available Pu dissolves.
- 77% of available fission products dissolve.
- Of the off gas available for release to off gas system, 100% is released.
- Excess acid added is twice as much as required by stoichiometry.
- UO₂ and U₃O₈ have the same solubility.
- Actinides (bar U and Pu) dissolve in the same manner as Pu.
- MO₂ FPs assumed to dissolve in the same manner as U.
- All NO_x generated is released to off gas.
- Acid concentration is set to adjust so that U conc in product liquor is ~100g/l ~8M.
- As much O₂ is added as required by the reactions – no excess assigned.
- Lanthanides dissolve in the same manner as FPs.

11.2.3. ACID WASH

This model represents the acid wash units which will wash solids from the centrifuges. This model has two instances – one after each centrifuge. The inputs to this sub-model are the solids from the centrifuge and acid, its output is a solid-rich slurry.

- The pull of acid is determined by a solid-liquid ratio.

11.2.4. DENSITY MODEL (AQ, GAS, OR)

There are three density sub-models which work within the model, one for the aqueous and organic phases, and one for gas phases. Where streams contain solids only, no density calculation was used resulting in no value for the volumetric flow.

Aqueous Density Assumptions

- Temperature is constant and fixed to that of the entire plant (assumed as 30°C).
- NNL's BRADSIM model for density calculation is used here.
- The effects of pressure are negligible.
- Minor actinides and lanthanides are regarded the same as U and Pu as their effects are small in comparison.
- Only cladding is taken into account when accounting for density differences due to solids – this is because it is the main contributor of solids throughout the process.
- The density is based upon the concentration of nitric acid and then corrected based on the quantities of heavy metal dissolved within it – all other dissolved species are assumed to have a negligible effect on the density.

Organic Density Assumptions

- Temperature is constant and fixed to that of the entire plant (assumed as 30°C).
- NNL's BRADSIM model for density calculation is used here.
- The effects of pressure are negligible.
- The effects of pressure are negligible.
- OK density is 778 kg/m³.
- Minor actinides and lanthanides are regarded the same as U and Pu as their effects are small in comparison.
- The density is based on the solvent, the quantity of extractant and acid within the solvent, and the quantity of heavy metal, no solids have been taken into account.

Gaseous Density Assumptions

- Temperature is constant and fixed to that of the entire plant (assumed as 30°C).
- Gases are assumed to obey the ideal gas law and thus the ideal gas law equations can be used here.
- Pressure for the system is fixed at atmospheric pressure.
- Average molecular weights were used in conjunction with the ideal gas law.
-

11.2.5. BUFFER TANK

This sub-model is a simple flow in/flow out unit designed to act as a break between two units or to act as an addition point where two or more streams meet, and combine before passing to the next unit.

- The effects of time and residence time are neglected.
- The effects of mixing are neglected.

- There is no interaction between the species (i.e. no reaction) whilst within the unit other than the combination of the streams.

11.2.6. CALCINE

This model represents the calcine and drying stage of the mixed actinide finishing line. The inputs to the model are the filtered product and oxygen inlet. The outputs of the model are the mixed actinide product and off gas treatment. Reactions occur within this unit.

- All available actinides react.
- Inert gases will feed the calciner to prevent the absorption of moisture however this stream has not been modelled.
- All actinides, U and Pu, are regarded as the same in terms of chemical reaction.
- Off gas consists only of O₂, CO₂, CO, and H₂O – entrainment of solids has been neglected.
- Actinides are perfectly dried.
- No gas is entrained with the product powder.

11.2.7. CENTRIFUGE

There are two instances in which the centrifuge model is used, after each NO_x conditioning step. This model represents the separation and concentration of solids from the main process stream. The feed is dissolver product liquor and the outputs are solids and clarified liquor.

- All solids (Cladding and insoluble) exit the unit via the solid stream – no solid entrainment.
- A fraction of the liquor is entrained with the solid stream.
- Semi-volatile fission products are assumed to be insoluble in nitric acid and therefore follow the solids.
- For the centrifuge following the Ag dissolver - a solid fraction value of the centrifuge cake is calculated based on the mass of U and Pu (in liquor form) compared to the cladding and insoluble fission products. This value is then used to calculate the liquor entrainment. The liquor entrainment is then applied to all appropriate compounds.

11.2.8. EVAP (U FINISHING)

This Sub-model represents the U evaporator in the U Finishing section of the plant. The quantity of water evaporated is based on the quantity of uranium which exits the unit. The inputs are the product-bearing liquor from chemical separation and the outputs are evaporator condensate and concentrated liquor to thermal denitration.

- Off gas is condensed completely before exiting the unit – the unit encompasses the condenser.
- No non-volatile species are lost to effluent.
- Only water, nitric acid and solvent are lost to effluent.
- All solvent is lost to effluent – there is no solvent entrainment in the concentrated product stream.

- The extent of evaporation is set such that the concentration of elemental U is set at 1000gU/l in the concentrate stream. A set evaporation ratio could have been used but due to the excess water in the system, the quantity of product would have been incorrect. The water driven off as condensate is indicative of the scale of the excess water within the system caused by the factors used from the hot tests.

11.2.9. EVAP_OX (MA FINISHING)

This model represents the conditioning stage prior to oxalate precipitation (In MA Finishing) where the feed is concentrated. The input is the mixed actinide stream from chemical separation; the outputs are condensate and concentrate which feeds the valency conditioning unit.

- The unit encompasses a condenser so all off gas is condensed prior to leaving the unit.
- A fraction of the plutonium reacts with the water converting from Pu^{4+} to Pu^{6+} .
- No actinides, other than Pu change valence.
- A fraction of the nitric acid and nitrous acid is routed with the evaporated water in the condensate.
- No other components except nitric acid, nitrous acid and water are routed to condensate.
- None of the solvents or additives are carried forward in the concentrate - representing the solvent destruction steam strip expected to precede evaporation.
- An amount of water is evaporated.

11.2.10. FUEL_FEED

This sub-model represents the fuel feed into the process. The mass composition of the fuel is pulled into this model along with the heavy metal throughput of the plant, the model then calculated the related total throughput, and the mass flow of each component.

- No reactions occur.
- Assumes heavy metal throughput comprises of actinides and lanthanides only.
- Assume nitrate form of actinides are $\text{Np}(\text{NO}_3)_4$, $\text{Pu}(\text{NO}_3)_4$, $\text{Am}(\text{NO}_3)_3$, $\text{UO}_2(\text{NO}_3)_2$.
- Lanthanides (and lanthanide nitrates) are modelled as a single compound with a weighted average molecular weight.

11.2.11. FUEL PREP (VOLOXIDATION)

This model is designed to represent the voloxidation stage within the head end section. The fuel self-pulverises and subsequently separates from the cladding. The inputs to the stage are the fuel feed and oxygen feed. The outputs are waste cladding, gases to off gas treatment and output to acid dissolver

- It is assumed that, for flowsheeting purposes, the cladding separates from the fuel at this point however, the design considered will involve the cladding separating at the acid dissolver. This is not expected to impact on the overall mass flows of species.
- A small fraction of fuel is lost to off gas as fines.

- There is no excess oxygen pulled into the system. The quantity of oxygen is exactly what is required according to chemical reaction.
- A fraction of volatile fission products are released to off gas.
- It is assumed that the normal operating temperature will be sufficient to volatilise all Cs.
- All iodine is released.
- All tritium is released.
- A fraction of the UO_2 available will react to form U_3O_8

11.2.12. MIXER (CDTA)

This sub-model represents the addition point of CDTA into the process – mixing it with the feed to the second cycle.

- The composition of the CDTA stream is calculated so as to adjust the output stream to 0.055M CDTA and 5.9M HNO_3 . It also dilutes the output of the mixer compared to the output of the 1st cycle.
- A dilution factor, based on hot test data, was applied to the mass flow of water to calculate the water associated with the aqueous CDTA stream [30].

11.2.13. NO_x _CONDITIONING

This model represents the NO_x conditioning stage after the dissolvers. The stage converts both Pu^{6+} and Np^{6+} to Pu^{4+} and Np^{4+} . This is carried out by reactions with NO_2 . The inputs to the model are the feed from the acid dissolver NO_x gas. The output of the model goes to the centrifuges for separation.

- Both Pu^{6+} and Np^{6+} are converted to Pu^{4+} and Np^{4+} through reactions with NO_2 .
- No side reactions occur.
- The NO_x is fed as required by the reaction stoichiometry; no excess is fed.
- A mass fraction of plutonium and neptunium react. Modelled as the same fraction.

11.2.14. OX _FILTRATION (MA FINISHING)

This model represents the filtration stage after oxalate precipitation. The inputs to the model are post oxalate precipitation reactor. The output is an effluent stream that goes to waste and a product stream that progresses to calcine and drying.

- The components separate based on the THORP flowsheet.

11.2.15. PRECIPITATION (MA FINISHING)

This model represents the oxalate precipitation reactor. The inputs to the reactor are a valence conditioned actinide feed and an acid feed containing oxalic acid and water. The output goes to the oxalate precipitation filter.

- All the available actinides react.
- All actinides are assumed to follow the same reaction scheme as Pu.
- The hydration number of the actinide oxalates will be the same as Pu.
- As much acid that is needed is drawn in as an input.
- Water is in excess - acid is fed at a given concentration, pulling water to match this concentration NOT what is required for reaction.

11.2.16. FIRST_1_CYCLE (PRIMARY SEPARATION)

This sub-model is designed to represent the Minor Actinide extraction stage of the first GANEX cycle. The inputs are the dissolved fuel feed and solvent (DEHiBA). The outputs are actinides and FPs fed to second GANEX cycle and U-rich solvent fed to U backwashing. No reactions occur - only solvent and aqueous extraction and backwashing.

- No reactions (just extraction).
- Assume carry over of acid into solvent phase and solvent into aqueous (via dissolution/entrainment) is 0.1%.
- The ratio of DEHiBA mass flowrate to heavy metal mass feed rate being the same as in NNL hot tests [30]
- All other solvent components are in mol ratio to DEHiBA, except the diluent (OK), which is adjusted by density of the stream being set.
- The ratio of HNO₃ mass flowrate in the scrub to heavy metal mass feed rate being the same as in previous models [2] This is based on the product of the heavy metal throughput and a HNO₃ ratio (based on hot test data [30]).
- All other aqueous components in the scrub feed are in mol ratio to the HNO₃ except the diluent (in this case water), which is adjusted by density of the stream being set.
- Densities are calculated from the Aq_densityModel and Or_DensityModel.
- Cm is handled via a decontamination factor (DF) and has been assumed to behave like Am.

11.2.17. FIRST_2_CYCLE (U BACKWASH)

This model is designed to represent the U stripping in the first cycle GANEX process. The inputs are the U-rich solvent and nitric acid. The outputs are feed to solvent recycle and u product.

- No reactions occur (just extraction).
- Assume carry over of acid into solvent phase and solvent into aqueous (via dissolution/entrainment) is 0.1% - value is unknown at present.

- The volume of aqueous fed to the backwash is the same as the volume of solvent fed to the backwash, i.e. the ratio of aq volume to solvent volume = 1. This is slightly pessimistic. In practice, because there is so little lanthanides in the stream it is possible that a much lower ratio could be used, however this would have to be weighted up against potential maloperations, such as if U breakthrough occurred upstream and there was too little aqueous in the backwash to accept it then significant amounts of crud could form in the backwash.
- All other aqueous components in the scrub feed are in mol ratio to the HNO₃, except the diluent (in this case water), which is adjusted by density of the stream being set.
- DFs are taken from previous model work [2].
- Densities are calculated from the Aq_densityModel and Or_DensityModel.
- Cm is handled via a decontamination factor (DF) and has been assumed to behave like Am.

11.2.18. SECOND_1_CYCLE (TRU SCRUB)

This model is designed to represent the fission product extraction stage of the second GANEX cycle. The Inputs are the aqueous stream from the first GANEX cycle, nitric acid and solvent feed. The Outputs are an Actinide and lanthanide aqueous stream that enters the next stage of the GANEX process and a fission product waste stream. No reactions occur - only solvent and aqueous extraction and backwashing

- No reactions (just extraction).
- Assume carry over of acid into solvent phase and solvent into aqueous (via dissolution/entrainment) is 0.1% - value is unknown at present.
- The ratio of TODGA mass flowrate to heavy metal mass feed rate being the same as previous models [2]. Based on Hot Test Data [30].
- All other solvent components are in mol ratio to TODGA, except the diluent (in this case OK), which is adjusted by density of the stream being set.
- The ratio of HNO₃ mass flowrate in the scrub to heavy metal mass feed rate being the same as previous models [30].
- All other aqueous components in the scrub feed are in mol ratio to the HNO₃, except the diluent (in this case water), which is adjusted by density of the stream being set.
- DFs are the same as in previous models [2].
- Densities are calculated from the Aq_densityModel and Or_DensityModel.
- Cm is handled via a decontamination factor (DF) and has been assumed to behave like Am.

11.2.19. SECOND_2_CYCLE (ACTINIDE STRIP)

This model is designed to represent the actinide strip stage of the second GANEX cycle. The Inputs are the solvent feed containing An and Ln from the first cycle of the GANEX process, an aqueous acid feed and a solvent. The Outputs are actinides rich aqueous stream and a lanthanide-rich solvent stream. No reactions occur - only solvent and aqueous extraction and backwashing

- No reactions (just extraction).
- Assume carry over of acid into solvent phase and solvent into aqueous (via dissolution/entrainment) is 0.1% - value is unknown at present.

- The ratio of TODGA mass flowrate to heavy metal mass feed rate is the same as in previous models based on Hot Test Data [2] [30].
- All other solvent components are in mol ratio to TODGA, except the diluent (in this case OK), which is adjusted by density of the stream being set.
- The ratio of HNO_3 mass flowrate in the scrub to heavy metal mass feed rate being the same as in previous models [2] [30].
- All other aqueous components in the scrub feed are in mol ratio to the HNO_3 , except the diluent (in this case water), which is adjusted by density of the stream being set.
- DFs are the same as in previous models [2].
- Densities are calculated from the Aq_densityModel and Or_DensityModel.
- Cm is handled via a decontamination factor (DF) and has been assumed to behave like Am.

11.2.20. SECOND_3_CYCLE (LANTHANIDE STRIP)

This model is designed to represent the lanthanide strip of the second GANEX cycle. The inputs are a lanthanide-rich solvent stream and an aqueous stream. The outputs are a lanthanide-rich aqueous stream and a solvent raffinate. No reactions occur - only solvent and aqueous extraction and backwashing

- No reactions (just extraction).
- Assume carry over of acid into solvent phase and solvent into aqueous (via dissolution/entrainment) is 0.1% - value is unknown at present.
- The ratio of TODGA mass flowrate to heavy metal mass feed rate being the same as in previous models [2] [30].
- All other solvent components are in mol ratio to TODGA, except the diluent (in this case OK), which is adjusted by density of the stream being set.
- The volume of aqueous fed to the backwash is the same as the volume of solvent fed to the Ln Strip, i.e. the ratio of aq volume to solvent volume = 1. This is slightly pessimistic.
- All other aqueous components in the scrub feed are in mol ratio to the HNO_3 , except the diluent (in this case water), which is adjusted by density of the stream being set.
- DFs are taken from previous models [2].
- Densities are calculated from the Aq_densityModel and Or_DensityModel.
- Cm is handled via a decontamination factor (DF) and has been assumed to behave like Am.

11.2.21. SHEARING

This sub-model represents the mechanical shearing of fuel feed

- No fuel is lost to off gas as fines in this unit.

11.2.22. SOLVENT ADDITION (FIRST AND SECOND CYCLE)

This sub-model represents the fresh solvent addition that replaces the solvent that is purged off in the solvent recycle stage. The inputs to the model are purged solvent (containing actinide and misc. compounds) and fresh solvent. The output of the model is the solvent feed to the GANEX solvent extraction cycles. Two instances of this model exist for the first and second cycles.

- Fresh solvent contains no impurities.

11.2.23. SOLVENT RECYCLE (FIRST AND SECOND CYCLE)

This sub-model represents the purge stage of the solvent recycle in the two GANEX cycles – it appears as two instances. The input to the unit is the solvent from the GANEX cycles. The outputs are the solvent to the solvent addition stages and a purge stream. The purpose of the model is to remove dirty solvent in the purge stream (clean solvent is added downstream). The model does not act as a true solvent wash; it simply has a purge stream which removes an amount of contaminants based on an assigned DF.

- At present the unit is assumed to operate continuously with a purge and top up with fresh solvent. The amount of HM/FPs/aqueous products that build up is unknown so it is assumed that the solvent recycle removes a 1% purge. This is pessimistic with regard to the contactor sizes since it is a low removal rate, so there is a higher build-up of metals and thus larger flowrates are required. Although the effect is small since the recoveries of the various species are good.
- Densities are calculated from the Aq_densityModel and Or_DensityModel.

11.2.24. TDN_SYSTEM

This model represents the thermal denitration system of the uranium finishing line including the air pre-heat, TDN reactor and overhead condensation. This model does not consider condensation as a separate entity. The UN thermally decomposes to O_2 , NO_2 and NO . The input ports are fluidising oxygen and the concentrated uranium feed. The outputs are uranium product powder and a condensate stream - air is routed with the condensate stream. This could be better represented using a separate output stream for off gas.

- Some uranium is lost to the off gas.
- Water in the product is calculated based on a ratio.
- No gases are entrained with the products.
- Assumed NO , NO_2 and O_2 are generated in equal measures.
- All available uranium is assumed to react.
- There is a requirement for air to fluidise and heat the product for thermal denitration to occur. This is calculated using an air_to_u_ratio.
- All air is assumed not to be consumed in any reactions and is routed with the condensate rather than a separate off gas stream.

11.2.25. VALENCY_CONDITIONING

This model represents the valance conditioning stage of the oxalate precipitation finishing line. The inputs are the feed from chemical separation and an acid feed. The outputs feed the precipitation reactor

- All actinides are conditioned to Ac 4+.
- Unknown mechanism for reactions - to mass balance unit, a H₂O₂ stream was added - the exact mechanisms of what will occur here is unknown. It has been speculated that like THORP, due to the high activity and high Pu concentrations radiolytic reactions will convert all Pu to Pu(IV) without the need for conditioning. THORP originally had H₂O₂ added but, once the discussed radiolytic reactions were found to be sufficient for conversion, the conditioning vessel in THORP was removed from use.
- Conversion of all actinides other than U and Pu are the same.
- All gaseous products are removed via an off gas system, all liquid products are routed to the next unit operation.

Assume H₂O₂ is not in excess - mechanism is not known and it has been included only for mass balance purposes.