High Level Waste Processing in the U.K. – Hard Won Experience that can Benefit U.S. Nuclear Cleanup Work

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ABSTRACT

High level liquid waste (HLLW) is generated as a by product of spent nuclear fuel reprocessing. Almost 2 billion curies of high level waste (HLW) have been generated in three generations of reprocessing plants at the U.K.'s Sellafield site. The waste management strategy is based upon minimizing the final volume of the HLLW arising by retaining an acidic flowsheet, segregating the less active salt-containing streams where possible, and only neutralizing the very low activity wastes. The treated HLLW waste is vitrified and the less active material encapsulated in cement.

Irrespective of the waste stream, the liquid waste facilities comprise buffer storage for the dilute arisings, evaporation plant; concentrate storage and finally treatment facilities with associated product stores. The equipment utilized for the HLW processing is described in detail and the design principles and key design features to ensure safe, efficient and continued operation are discussed. Extensive R&D programs have invariably underpinned equipment and process selection and several examples are presented.

Solutions to the specific challenges presented by removal of fission product decay heat, agitation of suspended solids, blockages from suspended solids, corrosion, gas clean up, representative sampling, and potentially very hazardous fault conditions have been developed and are described. Over the past 55 years extensive operational experience has been gained, sometimes the hard way. Several of the key lessons learned are discussed, with particular emphasis on requirements to assure facility operability and maintainability, throughput, and adequate or extended service life. The importance of system design and adequate testing is emphasized. The value of excellent feedback between operators and designers and having an integrated approach to R&D, design, construction and commissioning is demonstrated.

INTRODUCTION

The UK has reprocessed irradiated nuclear fuel for over 50 years for both defense and civil purposes. The majority of the U.K.'s reprocessing has been conducted in three generations of reprocessing facilities at the Sellafield Site in the northwest of England, all of which have utilized solvent extraction to separate uranium and plutonium from each other and from the fission product waste. In the U.K. the radioactive liquid waste streams are classified and subsequently treated according to the level of radioactivity contained. They are broadly characterized as high, medium, and low active.

The high level liquid waste (HLLW) principally comprises the acidic aqueous raffinates from the primary separation stage of the reprocessing solvent extraction processes. HLLW from both the civil and military processes is stored in concentrated acidic form in stainless steel tanks prior to vitrification. Medium active liquid waste (MALW) streams are also stored in concentrated form prior to treatment comprising precipitation and filtration of the resulting solids phase. This treatment is primarily aimed at removing the actinide constituents but also serves to significantly reduce the fission product content.

The constituents of the Sellafield HLLW and MALW are similar to those in the high level waste currently stored in alkaline form in carbon steel tanks at the Hanford and Savannah River sites in the U.S.A. A key difference in the U.K. waste management strategy is that the wastes are processed and stored in the acidic form. This enables the total volume of HLLW to be minimized thus significantly reducing the number of tanks required for storage. It also means, however, that only high integrity stainless steel plant and equipment can be used.

In this paper the strategy for management and disposition of the above waste streams is presented and discussed. The facilities, systems, and processes utilized for the treatments of the HLLW streams are described and the key operational and technical challenges, together with their respective solutions, are also discussed. Where appropriate, usually for comparison purposes, reference is made to the MALW treatment facilities and processes.

NOMENCLATURE

AGR	Advanced Gas-Cooled Reactor
ALR	Air Lift Recirculation system
AVH	Atelier Vitrification La Hague
BWR	Boiling Water Reactor
BNFL	British Nuclear Fuels plc, parent company of British Nuclear Group
BNG America	British Nuclear Group’s U.S.-based subsidiary
EARP	Enhanced Actinide Recovery Plant at Sellafield
ESP	Electrostatic Precipitator
FSIF	Full Scale Inactive Facility (vitrification full scale non-radioactive pilot plant)
HLLW	High Level (radioactivity) Liquid Waste
HLW	High Level (radioactivity) Waste
HALES	Highly Active Liquor Evaporation and storage plant at Sellafield
HAL	Highly Active Liquor (liquid)
HEPA	High Efficiency Particulate Arresting filters
LWR	Light Water Reactor
MAC	Medium Active (liquid waste) Concentrate
Magnox	Uranium metal fuel clad in magnesium-aluminum alloy
MALW	Medium Active Liquid Waste
NAG	Nitric Acid Grade stainless steel
PJM	Pulse Jet Mixer
RFD	Reverse Flow Diverter: a type of fluidic-operated liquid pump
SEC	Salt Evaporator Concentrate
TBP	Tri-Butyl Phosphate (mixed with kerosene for use as the solvent extraction solvent)
Thorp	Thermal Oxide Reprocessing Plant at Sellafield
VOSL	Vacuum Operated Slug Lift, a type of vacuum assisted airlift
WVP	Windscale Vitrification Plant at Sellafield

LIQUID WASTE STREAMS

The high level liquid waste contains the bulk (>99%) of the fission products from the reprocessed fuel and is predominantly composed of the aqueous raffinates from the primary solvent extraction separation stages. Various acidic medium active streams are also directed to the HLLW route.

Approximately 6 to 9 m^3 (1600 to 2400 US gallon) of unconcentrated HLLW is generated for each metric ton of uranium processed and typically the specific activity of this stream is 0.4 to 4.0E15 Bq/m^3 (41 to 409 Ci/US gallon). The HLLW stream is heat generating which drives several requirements in the design of the Highly Active Liquor Evaporation and Storage (HALES) facilities and the Windscale Vitrification Plant (WVP). These are discussed more fully below.

The unconcentrated medium active waste streams, which are aqueous raffinates from the secondary stages of separation, the purification cycles, and solvent wash, typically have a specific activity in the range 0.004-0.4E10 Bq/m^3 (0.04 to 4.1E-4 Ci/US gallon) (and are therefore not self heating even when concentrated). The total arisings are measured in several tens of cubic meters per day prior to evaporation.

In the most modern reprocessing facility, the Thermal Oxide Reprocessing Plant (Thorp) [1], which processes the significantly more highly irradiated AGR, LWR and BWR uranium oxide fuel, the flowsheet uses "salt-free" valency adjustment reagents that do not add extra cations to the process. The concentrated salt-free medium active liquid effluents can therefore be combined with the HLLW arising prior to dispatch to the vitrification plant. This is in contrast to the situation in the Magnox (metal fuel) reprocessing facility, where the MALW stream is concentrated and stored in dedicated facilities.

In both Magnox and oxide fuel reprocessing, the medium-active salt-bearing streams are segregated from the salt-free streams. All of the MALW streams, whether salt-bearing or not, are concentrated in dedicated evaporation facilities. Originally the intention was to delay-store the medium active Magnox streams prior to sea discharge but the site discharge limits, particularly for the actinides, were significantly reduced necessitating treatment of those concentrates.

The Medium Active Concentrate (MAC and SEC) is then processed through the Enhanced Actinide Removal Plant (EARP) where the actinides are precipitated out, filtered using ultrafilters, and encapsulated in cement. As mentioned above the only exception to this is the salt free MALW arising in Thorp. That stream is routed to the HALES plant and is eventually vitrified.

OVERVIEW OF THE WASTE TREATMENT FACILITIES

Irrespective of the waste stream, the facilities comprise buffer storage for the dilute arisings, evaporation plant, concentrate storage and finally treatment facilities with associated product stores.

Equipment and process selection followed extensive R&D programs, usually culminating in extended testing and inactive trials on full-scale pilot plants. Where appropriate, individual plant items were also tested, in full scale mock-ups using suitable waste simulates, prior to installation in the plants themselves.

In the case of the HLLW stream, because of the high specific gamma activity and radiation, the facilities are heavily shielded and designed to be maintenance-free wherever possible. If this is not so, as in parts of the vitrification facilities, then design features are incorporated to enable full remote maintenance to be performed.

The majority of the facilities in the HALES plants which are, or could be, in contact with the waste are therefore designed with no moving parts inside the secondary containment. Fluid energy devices are used for all liquor transfers, tank emptying and vessel agitation. These include [2] fluidic pumps, steam, air & water ejectors, air lifts, vacuum operated slug lifts (VOSLs), vacuum and pressure induced siphons. This design approach is also utilized in the liquor handling cells associated with WVP but clearly cannot be sustained in the vitrification and container handling cells.

Primary containment is provided by the vessels and pipework. These are located in massive, thick walled (typically 1.6m, 5.25ft) and reinforced concrete cells which also provide the secondary containment. The cells are either partially or fully lined with stainless steel cladding fitted with drainage sumps equipped with level monitoring, sampling and emptying facilities. Most cells are fitted with washdown sprays for the stainless steel cladding. All services to the vessels and the cells are introduced through adjacent shielded enclosures, known as "bulges", or unshielded cabinets if there is no risk of activity backflow. Containment of the radioactive material is further enhanced by parallel ventilation systems. The primary containment is maintained at a depression relative to atmosphere of nominally 750 -1000 N/m^2 (0.11 – 0.15 lbs/in^2) by the 'vessel' ventilation system and the secondary containment at 250 – 400 N/m^2 (0.04 – 0.06 lbs/in^2 by the 'cell' vent system. The operating areas are also ventilated but are nominally at atmospheric pressure. Thus airflow is always from the outside atmosphere into the cell containment and from the cell into the vessels. The ventilation airflows are drawn through a range of gas clean-up equipment prior to discharge to atmosphere.

In-cell components are constructed of very high-grade stainless steel to provide resistance to corrosion, the principal vessels being 18/13/1 (Cr/Ni/Nb) (Type 316) and the ancillary tanks and equipment are fabricated from 18/8/1 (Type 304). In more recent years a subset of Type 304, designated 304L and known as ‘Nitric Acid Grade or NAG, has been used instead of 18/13/1. This is very low carbon content steel.

The plant design life varies according to intended service but in the case of the tanks is not less than 60 years. Extremely conservative assumptions regarding anticipated liquor corrosion rates were incorporated into the vessel design. Tank wall thickness measurements carried out on the older single shell tanks, which at the time had been in service 25 years, showed no measurable thinning of the walls. None of these tanks have leaked. Another key design feature in the High Level Waste facilities is provision of spare capacity and redundancy and diversity in the type of equipment and services deployed throughout the facilities.

In the Medium Active waste facilities bulk shielding requirements are much less and contact maintenance is performed on many systems. So, for example, conventional mechanical pumps are deployed and whilst duty and standby are supplied, redundancy and diversity is not generally required. Where stainless steel is required it is almost universally Type 304. Special steels/metals have been utilized in areas such as the tube sheets on the MA evaporators to secure longer service life. Ventilation system requirements are less in the MA facilities.

HLLW EVAPORATION & STORAGE

Fresh arisings from the first Sellafield reprocessing plant were either sent directly to evaporator feed tanks or buffer storage. The original buffer storage facilities comprised 8 relatively large, 1200m^3 (317,000 US gallons), unagitated stainless steel tanks and these are still in service today, which provides considerable operational latitude and flexibility. (In recent years four of the tanks have been used to delay-store Salt Evaporator Concentrate.)

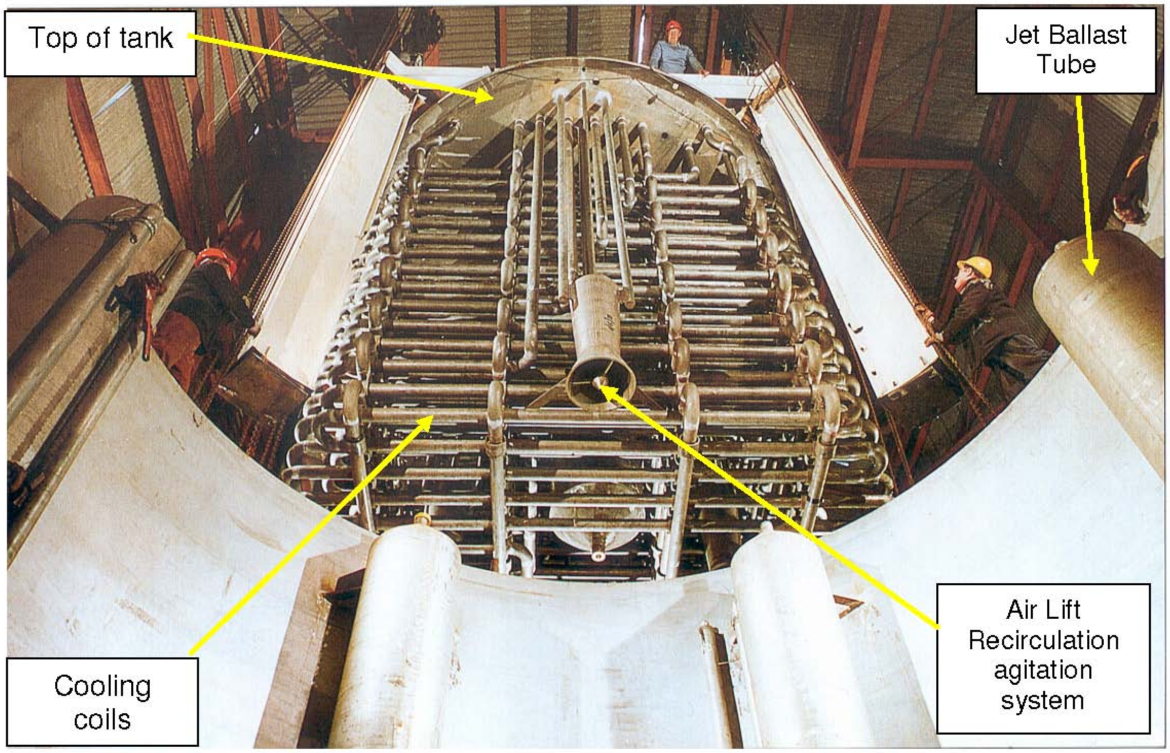

Fig. 1. HLLW storage tank under construction

Initially, when dealing with waste arising purely from the relatively small UK defense program, the storage strategy was based on storing moderately concentrated liquor in cooled but unagitated stainless steel tanks. The degree of concentration was primarily limited by the amount of solids produced, since the storage tanks were unagitated. These tanks are single shell, of cylindrical shape with the axis horizontal, with dished ends and approximately 70m^3 (18,500 US gallons) in capacity. Eight of these tanks were constructed, with 6 being filled and two retained as spares. Each tank had its own dedicated small evaporation unit.

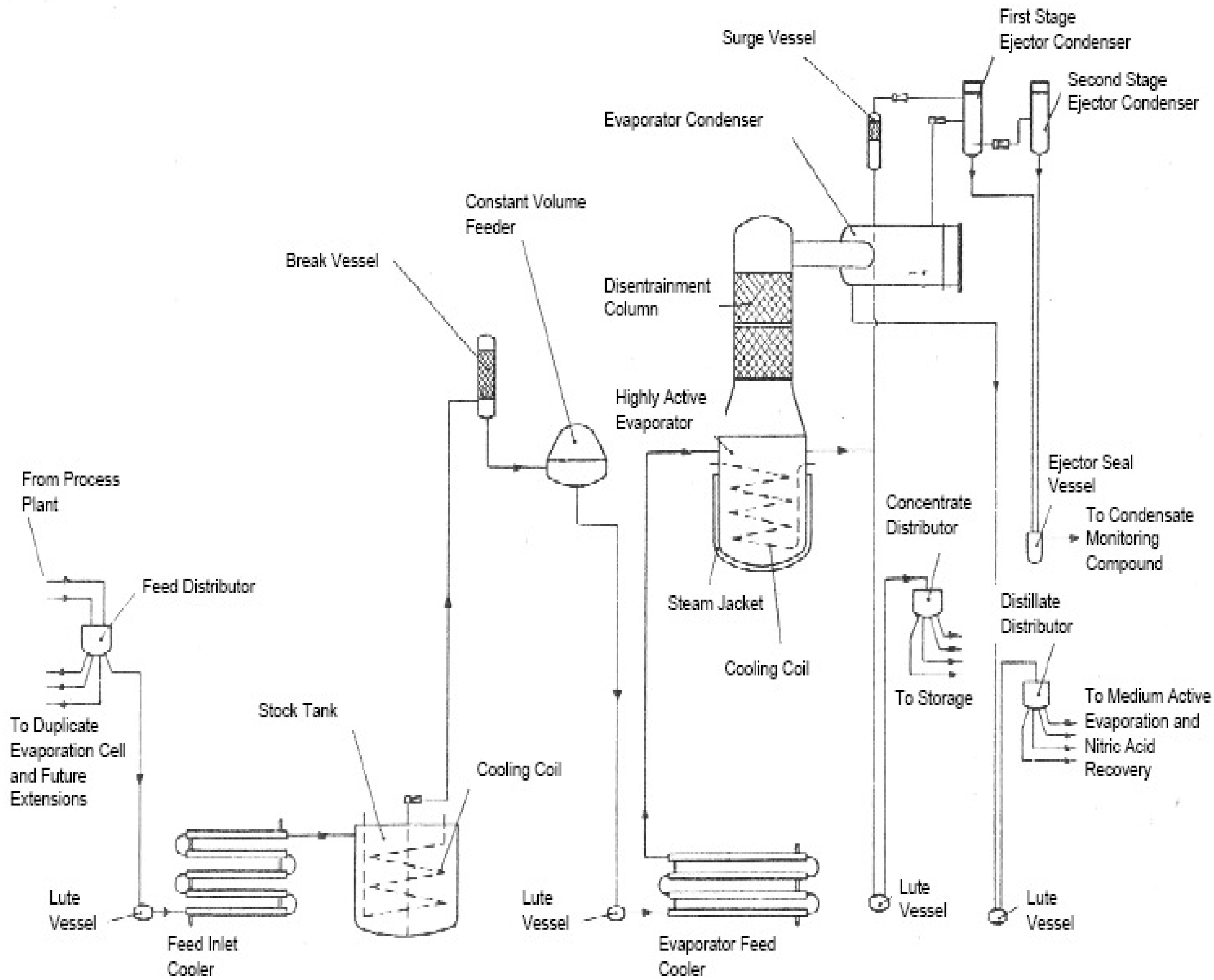

Fig. 2. Simplified flow diagram of HA evaporation plant

When commercial reprocessing of civil reactor was to be undertaken it was recognized that the HLLW facilities would also need to be significantly expanded, capable of dealing with the HLLW arising from the 1500 metric tons U/year Magnox reprocessing plant. New designs of both evaporators and storage tanks were therefore developed. After extensive full scale testing, the first tranche of the 'new' HLLW evaporation and storage facilities were constructed and brought into service from 1965 through 1970. Subsequent additions through 1990 have retained the same basic design, with various refurbishments and upgrades being implemented since then. All of the HLLW arising from reprocessing over 50,000 metric tons of spent fuel has been safely stored in either the old single shell tanks or the new double shell tanks

The 'new' design of storage tank is fabricated from stainless steel, is of cylindrical shape but with the axis vertical, has a flat bottom, is jacketed on the base and side walls, has multiple cooling coils, and two independent, diverse, agitation systems. Its capacity is nominally 150 m^3 (~40,000 US gallons). A total of 13 such tanks were built, with some being designated as spares and therefore unused. Fig. 1 shows a 'new' HLLW Storage Tank under construction, with the assembly of tank top, cooling coils and agitation systems ready to be lowered into the tank body.

Three evaporators, Units A, B, &C, are in use. Units A&B were commissioned in 1965 and Evaporator C was brought on line in 1990. Construction of a fourth unit is currently being considered. Both A&B have undergone a major refurbishment. The evaporators are simple kettle units. They are jacketed and A&B each have four coils. Evaporator C has six coils. A simplified flow diagram of the current generation of evaporators is depicted in Fig. 2.

When permission was given in the mid 1970s for the second commercial reprocessing facility, Thorp, to be built, a condition of the approval was that 'BNFL provide suitable vitrification facilities in order to minimize the amount of HLLW requiring to be stored on site and to convert the liquid waste into a form suitable for return of that waste to the country of origin of the spent fuel reprocessed'. Hence no storage tanks have required to be constructed since 1990.

Fig. 3 shows the total amount of waste, in curies, that is currently held in storage at the Sellafield Site either in liquid form or as vitrified solid waste. For comparison purposes, the total curies stored in liquid and vitrified form at the US Department of Energy Savannah River and Hanford sites are also shown.

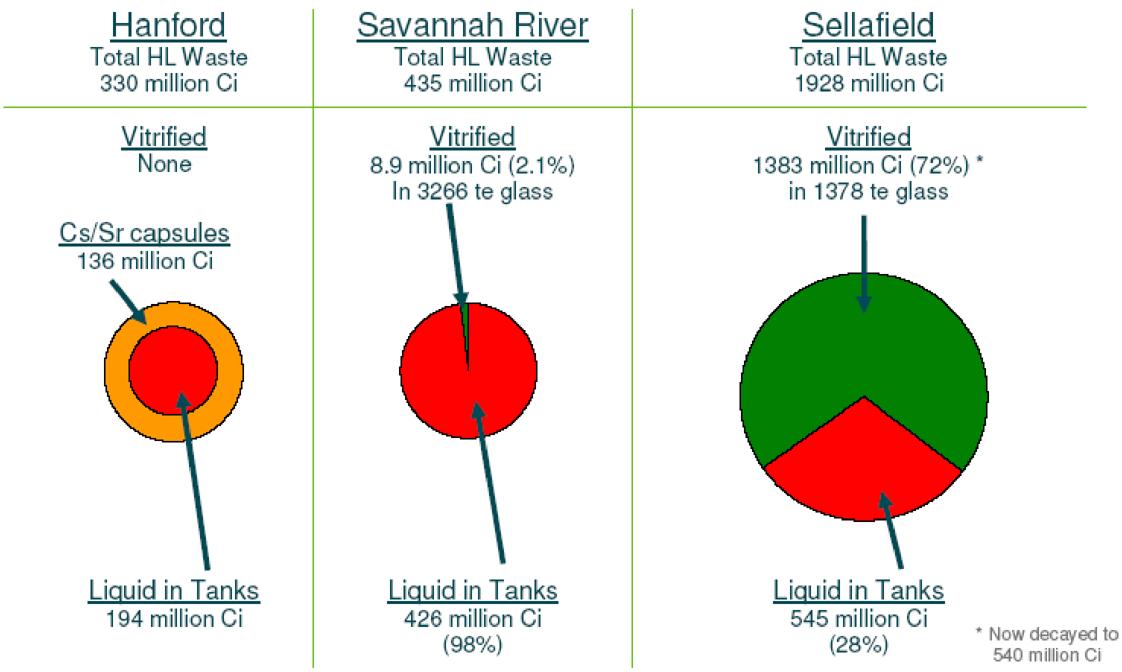

Fig. 3. Total curies of waste stored at Sellafield Site compared with the U.S. Hanford and Savannah River sites

SPECIFIC DESIGN FEATURES

The HLLW Evaporation and Storage plant is required to provide safe long term storage for the high level liquid waste until it is vitrified. In addition to shielding and containment, the facilities must deal with the challenges presented by:

- Removal of fission product decay heat,
- Agitation of suspended solids,
- Potential blockages from suspended solids;
- Corrosion,
- Disentrainment of radioactivity from exhausted air streams,
- The requirement to take representative samples for both process control and inventory purposes,
- Hazardous fault conditions.

The discussion in the rest of this paper relates only to the current generation of evaporators and the 'new' tanks. The original evaporators were taken out of service and the original tanks have either been emptied or are being blended with newer arisings prior to being fed to the Vitrification plants. Where relevant, reference is also made in this section to equipment and experience within the HLLW tank systems in the Highly Active Liquor (HAL) cells in the Vitrification plant.

The evaporators operate under reduced pressure and therefore reduced temperature in order to minimize corrosion rates. No evaporator has leaked. The evaporators are steam-heated, water-cooled when required and operated at relatively low boil-up rates to minimize entrainment into the condensate. Decontamination factors in excess of 1.0E12 from feed to condensate have been achieved with only simple disentrainment columns containing Lessing rings or simple baffle plates. The condensate or recovered nitric acid is eventually recirculated into the process.

The acidic feed to the HLW evaporators contains over 99% of the fission products from the spent fuel, corrosion products (e.g. iron, alloying constituents from the fuel rods such as aluminum), and phosphates from radiolytic breakdown of the Tri-Butyl-Phosphate (TBP) solvent used in the separation process. On evaporation, nitric acid is removed. Solids such as molybdenum oxide, zirconium phosphate and molybdate, and various phospho-molybdates, separate as the concentration is increased. Thorp arisings also contain barium-strontium nitrate. Because the solids could cause blockages, their quantity is carefully monitored and controlled, both in the evaporation plant and upstream and downstream. Zirconium molybdate is very difficult to remove if it plates out on hot surfaces and thus is a particular risk in the evaporators and the WVP primary dust scrubbers. The HLLW feed stream to the evaporators is steam-stripped to remove traces of the TBP and kerosene solvent prior to discharge from the reprocessing plants. This has the dual benefit of reducing the risk of "red oil" nitric acid-solvent reaction in the evaporators and of limiting the amount of solids precipitated when concentrated. In the Thorp facility the insoluble dissolver fines (that arise from the more highly irradiated oxide fuel) are also removed by centrifugation.

As a final stage in the evaporation the acidity is reduced by boiling down with water. (A variant on this process uses formaldehyde). Nitric acid is destroyed with no increase in the salt content of the wastes. The purpose of this is twofold: firstly, to reduce the rate of corrosion; and secondly, to increase the evaporation factor achievable before crystallization occurs. Typically the achieved evaporation factor is about 100 for Magnox fuel raffinates and can be as low as 50 for oxide fuel raffinates. Evaporators A&B can process about 65 m3/day (17000 US gallons/day), whereas Evaporator C with its additional coils can evaporate 95 m3 /day (25000 US gallons/day).

Post evaporation the concentrated liquor is transferred by siphon in batches to the storage tanks. The liquor specific gravity is nominally 1.4, but this is subsequently increased to about 1.55 through in-tank evaporation.

Removal of Fission Product Decay Heat

The HLLW storage tanks are operated at about 55 -60ºC (130 -140ºF) and cooling is necessary to achieve this. The bulk liquid temperature is kept above 45ºC (113ºF) to minimize crystallization. If uncooled, the decay heat is such that the tanks would eventually boil and, for the shortest out-of-reactor cooling time fuel, boiling would occur within a few hours. Each tank is therefore equipped with 7 independent cooling coils. The jackets are also water-cooled and the base and wall jackets are each equivalent to one coil in terms of heat removal capacity. The total heat removal capacity of each tank is about 2 MW. The cooling coil assembly can be clearly seen in Fig. 1. The total volume occupied by the coils and other tank internal equipment is less than 5% of the tank capacity.

Heat removal efficiency is reduced if crystals deposit on the cooling coils and excessive suspended solids impede free convection, which can lead to hot spots occurring. Consequently tank chemistry, which requires a detailed knowledge of tank contents, is controlled to minimize the amount of solids present and ensure the crystallization point (largely set by the magnesium-rare earth-nitrate content) is not exceeded.

The nitric acid concentration and tank temperature are therefore maintained between limits that ensure the solubility products are not exceeded and crystallization is minimized, the corrosion rates are acceptable, and that tank emptying equipment can always operate.

Agitation of Suspended Solids

The quantity and nature of the suspended solids varies depending upon the actual conditions. The bulk of solids are zirconium phosphates that are only semi-gelatinous and very voluminous, low density precipitates, occupying 50-60% of the volume of a sample after 24 hours settling. They are easily suspended. Typically they amount to only 5 wt. %. A much smaller fraction comprises the more dense and rapid settling phospho-molybdates which are correspondingly more difficult to suspend. Thorp arisings also contain significant quantities of zirconium molybdate and very rapid settling barium-strontium nitrate.

The tanks are equipped with six peripheral and one central "Jet Ballast" agitation systems, which are immersed in the tank liquid and controlled to operate in sequence. Jet ballasts are agitation devices that operate by using compressed air to periodically blow the liquid in each jet ballast tube out through a nozzle at the base, creating a jet of liquid that swirls across the base of the tank. The amount and pressure of air used is limited by a reservoir and, together with a control system, this ensures that air does not "overblow" into the tank. The jets are angled so that the base area of the tank is completely swept, including the corner with the side wall. The function of the jet ballast tubes is simply to prevent solids from settling. Full-scale inactive testing, using conservative simulates, demonstrated the functionality of the system, including recovering from extended periods of inoperation and hence solids settling.

Each tank is also fitted with four air lift recirculation (ALR) units which use compressed air to create an air lift system within the tank. This provides a very vigorous agitation of the tank contents and, in conjunction with the jet ballast tubes, completely homogenizes the tank contents. The jet ballasts operate continuously to keep solids in suspension, and the ALRs are used immediately prior to tank sampling operations. Again the functionality and ability to homogenize the tank contents was demonstrated through extensive testing on a full-scale tank mock-up. Operation of the ALRs can lead to foaming, particularly with fresh arisings, and therefore the tanks are fitted with foam detectors.

Both the Jet Ballasts and the ALRs require a minimum amount of liquor to be present in order to operate efficiently. In more recent applications of fluidic mixing, Pulse Jet Mixers (PJM) [2] have been used which operate with a suck and blow cycle. PJMs can operate effectively with very low levels of liquor in the tank.

Occasionally hot spots have developed in operational tanks, indicating accumulation or agglomerations of solids, and these have been dissipated using the agitation systems or temporarily modifying the tank chemistry. The incidence of hot spots has been more frequent since processing of the more radioactive Thorp raffinates began in 1995.

A more energetic PJM was developed for the HAL feed tanks in WVP, achieving better than the required 95% homogeneity. Various tank aspect ratios (height/diameter) in the range 1.0 – 1.5 were tested successfully as part of a broader demonstration of the effectiveness of the pulse jet system. However, a conservative approach has been taken for critical applications, where the tank aspect ratios used in practice seldom exceed 1.1. Extended full scale inactive trials, using a vessel of the aspect ratio selected for the HAL feed tanks and fully replicated tank internals, with accurate waste simulates, were used to prove design performance. Plant operational performance has been trouble free; with tank samples taken from the WVP feed tanks and the upstream blending tanks in the HALES facility providing excellent corroboration. In addition, a new compressed air driven fluidic pump, the Reverse Flow Diverter (RFD) [2] was utilized, instead of steam ejectors, to avoid diluting the vitrification line feed. Pulse jet equipment and RFD fluidic pumps, developed from this application, are being supplied to the Hanford Waste Treatment Plant.

Potential Blockages from Suspended Solids or Crystals

Despite the use of the agitation systems and control of tank chemistry to limit the amount of solids and prevent crystallization, as described above, there still exists the potential for blockages in pipes, pumping equipment and instrument lines.

Techniques used to further prevent blockages include the use of water or acid drip feeds into key components such as the bubbler (pneumercator) level measurement dip pipes and submerged steam ejectors. A very small flow of water or dilute acid is allowed to drip into the device in question via its service connection e.g. the air pipe to the pneumercator or steam line supplying the ejector. Clean water builds up inside the pipe, floating on the more dense liquor to a level determined by the relative densities of the water and liquor, and flows out into the tanks thus keeping the pipe and its entrance clear. The amount of water necessary is inconsequential compared to normal losses through in-tank evaporation. If the device is used in service then a short period at higher water flow to the drip is used to re-establish the clean condition. The efficacy of the drip system and the ability to sustain and recreate the clean condition was demonstrated on a full-scale mock-up of the emergency emptying steam ejector and pipework used in the double shell tanks, under the full range of operating conditions.

Vulnerable areas such as lutes in lines are fitted with wash facilities that enable steam, air, water, acid or other chemicals to be added. Freeze valves, using liquid nitrogen to freeze a short section of the liquor in a pipe, are fitted to enable upstream or downstream sections to be pressurized to clear blockages should they occur. Some pipes identified as particularly vulnerable to blockage are fitted with coaxial nozzles to allow the pipe to be cleaned with water, steam or acid. Fluidic pumps such as RFDs that are not in use are operated in a low energy 'agitation' mode which keeps the trapped liquor moving without discharging into the delivery line. Operational regimes are established whereby standby systems are operated in rotation, lutes are washed out after transfers and primed with nitric acid before use.

From time to time blockages have nevertheless occurred. They have generally been easily cleared using the installed provisions and, on occasions, have necessitated a revision of the operating practice or special measures to prevent recurrence. One such instance was blockage in the HAL feed return line from the Vitrification cell to the HAL feed tanks in the Vitrification plant. The precise location and nature of the blockage was established using a camera inserted into the line and then it was dissolved away using ammonium carbamate.

THORP arisings have presented more challenges because of their higher solids content than Magnox liquors, with some blockage events occurring since the start of processing waste derived from oxide fuel. The total number of events is still low, that is in single figures, and no plant or equipment has been lost because of blockages in over 50 years of operations at the Sellafield HLLW plants.

Allowing for and Preventing Corrosion

In addition to making adequate allowance for general corrosion loss it is necessary to ensure that areas of weakness are eliminated. Weld-affected zones are particularly vulnerable and require 100% inspection during construction. End grain and potential crevices are eliminated wherever possible.

High quality stainless steel is used throughout. The principal vessels are constructed of very heavy 18/13/1 plate. The wall thickness is calculated on the basis of reducing to not less than 50% at the design life using very conservative corrosion data. As described above actual measurements taken after 25 years of service showed no measurable tank wall thinning. The tanks walls are 1.6cm (5/8 inch) thick. Thicker plate is used in the evaporators and for tank bases. Concentrate-bearing pipe and in-tank fixtures such as cooling coils are nominally schedule 80 with not less than +15% on the wall thickness.

All primary containment welds and welds in equipment within the primary containment are fully radiographed and there are no penetrations in the base or side walls of the highly active waste storage tanks. Extensive use is made of forgings for penetration nozzles, jet nozzles, RFD elements, steam ejector components and other tank internal fittings. Where end grain is exposed during fabrication it is covered by buttering with weld metal. All plant and equipment is passivated using nitric acid before being brought into service.

There have been, nevertheless, a small number of coil failures in vessels caused by waterside corrosion. This has caused pinholes in a few HAST cooling coils and HLLW has escaped in small amounts into the cooling system. A similar failure has occurred on one tank jacket. In each instance the cooling water gamma monitors promptly detected the activity, shut down the circuit and the contaminated water was diverted from the Cooling Water Delay Tank into the active effluent recovery route. The mechanism for this failure has been shown to be associated with layers of sludge deposited in the coils, which together with peroxides from radiolysis of water and chloride ion from ocean-derived salt, create conditions that favor pitting/crevice corrosion. The sludge has been shown to be a mixture of silica and mild steel corrosion products from the water supply system. The cooling systems are single circuit recirculated, with cooling effected in forced draft cooling towers, open to the atmosphere and thus enabling wind blown dust and ocean spray to be picked up at the coastal Sellafield site.

Historically some coils were operated at low flows, which is conducive to deposition of solid material. Operational regimes have been instituted whereby coils not required are purged, dried and isolated. Open recirculated cooling systems have, where practicable, been replaced with primary-secondary systems. This enables close control of the water chemistry in the primary circuit.

There has been no indication of any penetration of the evaporator vessels due to corrosion. Originally the evaporators were considered to be most vulnerable. One coil has failed in each of Evaporators A&B after about 20 years of service. The coils are used for both heating and cooling. In this case the failure site has been shown to be where the coil is exposed to the vapor phase and the mechanism of 'thin film' corrosion. Camera inspection has shown droplets of nitric acid falling onto the hot, exposed coil surface and boiling off. The coils use low-pressure steam and the surface temperature is approximately 130ºC (266ºF). Evaporator life has been found to be determined by the service life of the coils.

Disentrainment of Air Streams

A simple disentrainment column is located between the evaporator and the condenser. Formation of submicron particles or droplets is minimized by utilizing very low boil-up rates and low vapor velocities in the liquid in the kettle-type evaporators. The micron-sized droplets are thus amenable to removal by impact mechanisms, and adequate DFs have been obtained with both Lessing rings and "knitmesh" packing. Lessing rings are utilized because of the additional life that they offer, even though the height of the disentrainment section is slightly greater. Baffle plates are utilized in Evaporator C.

Clean-up of the vessel ventilation air is achieved by operation of an electrostatic precipitator (ESP) in series with a simple irrigated, packed column followed by a water jet 'wet' scrubber. The packed column had the dual purpose of disentrainment and dehumidification. The fission product decontamination factors, calculated by analysis of both liquid and gaseous samples, across the combination of the ESP and packed column, significantly exceed design requirements even under the most onerous conditions in the HLLW storage plant e.g. when the air lift recirculators or steam ejector transfers are taking place. DFs well in excess of 1.0E6 have been measured. Although not proven conclusively, it is believed that the droplets leaving the ESP still retain an electrical charge which improves the removal efficiency in the downstream packed column. The DF across the wet scrubber is typically 20 -50, but under certain circumstances DFs as high as 100 have been measured.

When the HALES facilities were first constructed the cleaned vessel ventilation air was then discharged via a 120m (400ft) stack to atmosphere. Shortly after the new tanks were commissioned, primary and secondary HEPA filters were added. More recently an additional caustic scrubber has been added to remove ruthenium (Ru-106). The combination of ESP and irrigated packed column proved so successful that it became the standard for vessel vent clean up in all new chemical plant facilities at Sellafield.

Cell air is extracted via primary and secondary HEPA filters.

Representative Sampling for Process Control and Inventory Purposes

All of the storage tanks are fitted with liquid sampling systems enabling samples to be drawn from different points in the tanks.

Liquid is circulated from the tank to a point below the shielded sample bulge using a vacuum operated slug lift (VOSL) or airlift system. The VOSL is a modified air lift system whereby air is injected into a riser pipe that runs up to a small break pot to which vacuum is applied. The vacuum produces a larger submergence of the air inlet than would otherwise exist and, at the correct airflow range (established in full scale inactive trials), this creates a continuous flow of slugs of liquor up the pipe, separated by slugs of air at greatly reduced pressure.

The riser pipework is arranged to run almost horizontally, but on a very slight upward incline, for several feet just below a "bulge" (shielded access point in the shielded cell). It then discharges into a small break pot from which it returns to the tank. A small bore needle runs from the bulge and just penetrates the 'horizontal' pipe. With no sample bottle on the needle, air is drawn into the VOSL system because the open end of the needle is still at atmospheric pressure. No liquor escapes. When the VOSL has been operated for a short period the sample bottle is placed on the needle, by piercing its rubber cap with the needle. As the first vacuum slug passes by the needle it creates a reduced pressure in the bottle. As the next liquid slug passes by it is then drawn up into the bottle. The bottle is then withdrawn either manually or via an autosampling system in the newer facilities, and the rubber cap self-seals the needle penetration.

Depending upon the nature of the sample requirements, the tank contents may first be homogenized using the air lift recirculators. The effectiveness of this system has been proven during operation of the facilities by taking and comparing samples from different points in the tank.

In the HALES plant, placing the sample bottle on the needle, posting the sample into a shielded container, changing needles, washing needles etc, is done manually by the use of remote, shielded manipulators. In later plants such as the Vitrification Plant and Thorp, the manual sample bulges were replaced with fully automated, appropriately shielded, systems known as 'Autosamplers', in which the sample bottle is delivered to the system and dispatched to the laboratory via a vacuum operated pneumatic transfer pipe. These fully automated systems, which have been used extensively for the past 15 years, successfully taking and transporting samples of all level of activity direct from plant to laboratory, are to be installed in the Hanford Waste Treatment Plant [3]. The Autosamplers can be configured to work with a variety of sample delivery systems including VOSLs and fluidic pumps, and proprietary sample collection systems such as "Isolok" ®.

Hazardous Fault Conditions

The principal fault conditions of concern in the high level waste plants are failure of the tank fabric and loss of cooling. A philosophy of defense in depth through provision of redundant and diverse systems has been developed to ensure more than adequate mitigation.

As described above materials and methods of construction are carefully chosen to ensure high corrosion resistance and high integrity of the tanks and their equipment. The structure is seismically qualified and the cells are designed to withstand small aircraft or large projectile impact. The tanks are jacketed. The secondary containment is lined in stainless steel, which drains to sumps that have high capacity emptying ejectors which discharge to separate breakpots. Provision is also made to isolate the cell ventilation and connect it to the vessel ventilation system in the unlikely, but not impossible, event that significant quantities of liquid were to arise in the cell sump.

One tank in every four is kept completely empty as a standby spare. Each tank is fitted with two pairs of emptying ejectors; one pair submerged, the other above the liquid level. Each ejector discharges to separate breakpots. Later tanks are also fitted with a pair of fluidic pumps which can operate at temperatures close to liquid boiling point. All of the liquor transfer equipment used in the HLLW plant was fully tested prior to installation, using simulated waste on full scale mock-ups in which all pipework was accurately replicated, over a range of normal and fault conditions. The equipment is periodically operated in-plant to demonstrate no loss of performance.

The provision to mitigate loss of cooling is extensive. There are 9 separate cooling components in each tank and at full flowsheet conditions this initially provided about 100% redundancy in heat removal capacity. As the liquids have aged, and heat generation decreased, this redundancy factor has significantly increased. Three separate cooling water supplies exist, the normal site supply, a back-up piped directly in from an inland lake, and a permanently piped supply from a local river. The cooling water system has two separate forced draft cooling towers, which are sited in separate locations. Each tower comprises two cells and each cell is capable of carrying full duty. Each tower has two pairs of pumps. There are three separate cooling water distribution mains; the normal, a guaranteed, and an emergency.

Normal electrical power is provided through two separate supplies, routed in from different locations, with duty/standby load spreading across separate boards. The site supply is backed by diesel and gas turbine generators and dedicated generators are located at the HALES facilities. Mobile generators can also be connected at the facilities. Guaranteed non-interruptible supplies are available from batteries.

In the event that all of this was unavailable then cooling water can be supplied on a once-through basis from dedicated fire tender pumps, maintained in permanent readiness, extracting from a second local river.

WASTE VITRIFICATION

The Windscale Vitrification Plant (WVP) at Sellafield, which is based upon the vitrification process developed in France for the La Hague Reprocessing Facility, originally comprised two lines with a common HAL feed cell at the front and a shared decontamination and single control cell at the back end. A third vitrification line was added to deal with the additional arisings generated by the oxide reprocessing facilities. The process includes separate calcination and vitrification steps in contrast to the single step approach adopted in the U.S.A. where water is driven off from the melter. Each vitrification line comprises a vitrification cell housing the calciner, melter, the various constant volume feeders, and the dust scrubber and condenser of the primary off-gas plant; a pouring and container handling cell, housing a carousel machine which positions the container below the melter, cooling stations, and the container lid welding machine. There is a separate breakdown and dismantling cell for maintenance operations. The breakdown cell contains equipment for cutting up failed process equipment such as melter crucibles, calciner tubes, dust scrubbers, condensers etc. The breakdown cell also contains a welding machine for closing waste disposal containers. All of the vitrification line cells are remote maintenance.

The primary aim of the vitrification process is to convert the HLLW solution into a mixture of oxides which are then reacted at high temperature with glass-forming materials to produce a vitreous product on cooling. The basic process steps are evaporation to remove water and free acid, calcination to decompose the mixture of nitrates either partially or completely to the waste oxides, and finally reaction in a melt of the waste oxides with borosilicate glass formers to incorporate them into the glass matrix.

In the Sellafield vitrification plants the evaporation and calcination steps are separated from the final reaction stage in order to separately optimize the calcination and melting processes. Before being fed to the vitrification plant the HLLW is fully characterized by chemical analysis and lithium nitrate is added. The lithium inhibits the formation of refractory oxides of aluminum, chromium and iron during the calcination stage. Fig. 4 shows the melter and calciner together and indicates the complexity, from a remote maintenance standpoint, of the installation.

A key aspect of the operating strategy for the Sellafield vitrification plants is to assure vitrified product quality through quality control of the feed materials and good control of the process. In this way the need for active glass sampling or destructive testing of the product containers can be avoided. The major objective of the Vitrification R&D program was to examine different glass formulations and waste compositions and produce process operating envelopes, within which the facilities could safely and easily operate, that would always result in 'good' glass that met the customers' vitrified product specification. The operating envelope was then tested on a full-scale non-radioactive pilot plant (the Full Scale Inactive Facility, FSIF) and subsequently demonstrated on the plant using accurate waste simulates before hot operations commenced.

Calcination

The calciner is a stainless steel tube supported on a 3% slope and rotated in an electrically heated furnace. The furnace has four separate zones capable of operating at temperatures up to 850ºC (1560ºF). The calciner is operated at reduced pressure (-100mmWG) and air tightness is achieved by graphite sealing rings located in each of the end fittings. The HLLW solution is introduced into the upper end of the calciner; flowing down the tube it is first evaporated and then denitrated, spilling out of slots in the lower end of the tube as a finely divided powder or calcine.

The physico-chemical characteristics of the calcine depend upon the waste composition and the amount of lithium added. The correct particle size distribution is ensured by both mechanical and chemical means. Caking of the calcine is prevented by the tumbling action of a heavy iron rabble bar in the tube. Reagents are also added with the HLLW feed which, under the action of heat, decompose and produce large volumes of gas. This action helps create a friable calcine. The use of sucrose for this purpose was pioneered on WVP. It has the added advantage in that it is a denitrating agent and suppresses ruthenium (Ru-106) volatility by destroying nitric acid in the feed.

Vitrification

The fusion of calcine with the base borosilicate glass takes place in a directly heated metallic induction furnace – the melter crucible. The furnace is elliptical in section and has four principal heating zones. The melter operates at a wall temperature of approximately 1100ºC (2012ºF). The crucible neck is linked to the calciner lower end fitting by a lead glass seal

The crucible is fitted with a drain valve in the base to allow tapping off of the product glass. The base glass is introduced through an air lock into the lower end fitting of the calciner and falls into the melter together with the calcine. The base glass melts and the liquid glass dissolves the calcine. During the dissolution and digestion of the calcine any residual nitrates are decomposed. After a predetermined time the product glass is drained off into a stainless steel product container. The furnace is designed to produce 25 kg (55lb) of product glass per hour with a typical waste loading of 25 weight per cent.

Product Container Handling and Storage

The product container is 42cm (16 inch) in diameter and 1.3m (4.3ft) high and holds about 400kg (880lb) of glass, corresponding to two melter discharges or 'pours'. After filling it is allowed to cool and then a lid is welded on. It is then decontaminated using either high-pressure water jets or mechanical, abrasive means or both. The containers are taken into a control cell where their external surface is swabbed. If the swab confirms that the container surface is clean the container is transferred to an adjacent product store. Each vitrified container typically contains the waste from 8 metric tons of Magnox fuel or 2 metric tons of LWR fuel.

The storage area is an indirect, natural convection, air-cooled store in which the containers are stacked up to 10 high in closed thimble tubes**.**

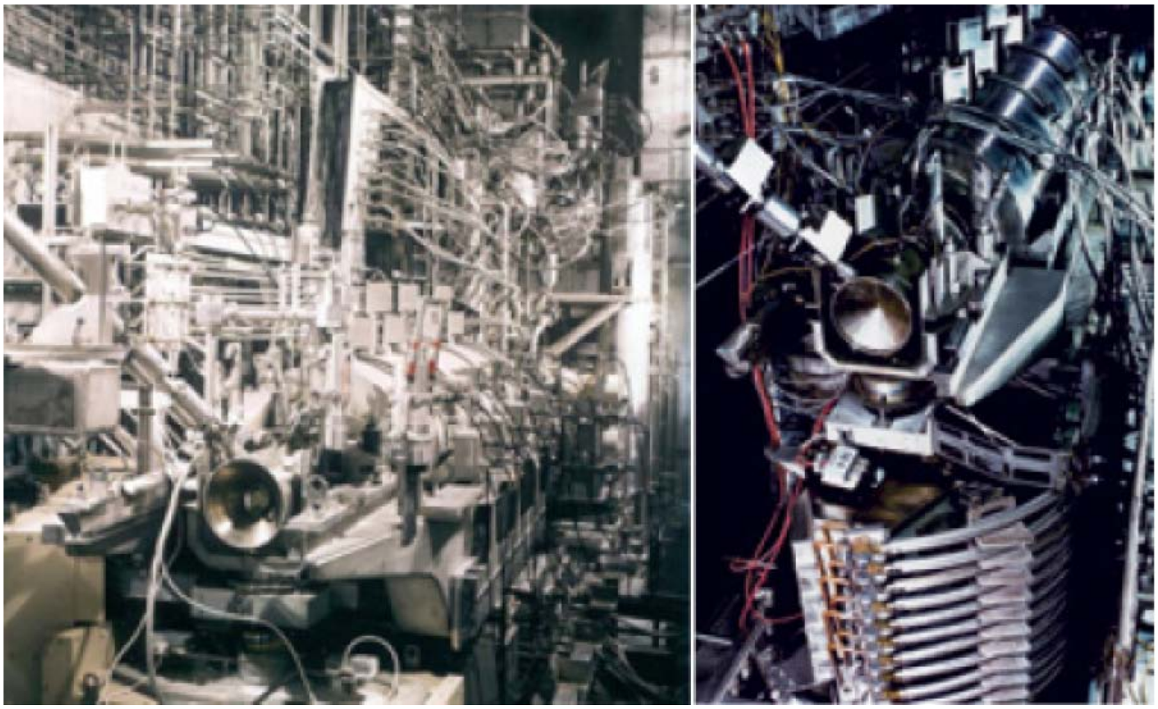

Fig. 4. View inside the WVP Vitrification Cell

Off Gas Treatment and Clean-Up

The primary off-gas system comprises a dust scrubber column, a tube condenser, and a bubble cap scrubber column for nitrogen oxide removal. Each vitrification line has a dedicated primary off-gas system. This is followed by a common secondary off-gas or vessel ventilation system which comprises duty and standby electrostatic precipitators, a wet scrubber, and primary and secondary high efficiency filters. The cell ventilation extract is discharged through primary and secondary filters.

The combined off-gas produced in the melter and calciner contains water vapor, nitrogen oxides, entrained dust and several radioactive species, but predominantly ruthenium (Ru-106) and cesium (Cs-137) volatilized by the process. The level of dust entrainment depends upon the temperature and chemical regime in the calciner, in particular the nature and composition of the HLLW feed together with any additives. It is typically 1%.

More than 99% of the material carried over to the off-gas is removed in the dust scrubber column. The highly active effluent arising from this operation is directly recycled to the calciner.

Maintenance

The HAL feed cell, as with all facilities in the Highly Active Liquid Storage and Evaporation Plants, is designed to be maintenance free, and the same redundancy and diversity of equipment with 'no moving parts' philosophy is deployed. These 'dark cells' are fitted with access ports through which cameras and inspection equipment can be installed.
This is not the case for the vitrification lines where the majority of in-cell equipment is remotely

replaceable using in-cell cranes, special tools and master-slave manipulators. The cells are fitted with windows and are permanently illuminated.

Prior to commencing hot radioactive operations, all of the installed equipment in the vitrification lines was fully remotely replaced, without operator intervention or presence in cell, using the installed maintenance tools and the draft operator and maintenance instructions. This rehearsal exercise was also fully repeated following the extensive thermal cycling that occurred during the inactive plant commissioning trials. These on-plant demonstrations of the full remotability of the vitrification lines were in addition to the extensive development and proving trials that had been done on full scale inactive equipment. Nevertheless several of the lessons learned from hot operation of Lines 1&2 and incorporated in Line 3 concerned improvements to facilitate remote maintenance. These are discussed more fully in the 'Lessons Learned' section below.

LESSONS LEARNED FROM VITRIFICATION PLANT OPERATION

Since commencing operations of the vitrification plants in 1990, a number of significant problems have been encountered and lessons have been learned, some of which have resulted in plant modifications and improvements. Others have meant that different operating regimes have been developed.

Considerable quantities of HLLW have been processed (Fig 3) and glass produced but initially the downtime between production runs was considerable, with the intricacy of the in-cell equipment significantly extending the time taken to effect both routine operations and remote maintenance.

Initially the key operational challenges and problems were:

- maintaining crane and master-slave manipulator availability,
- low melter life before replacement was necessary,
- accelerated calciner bearing wear,
- interlock control failures which resulted in significant operational downtime.

The less than expected lives of the melters, calciner bearings and some other in-cell components greatly increased the workload on the cranes, special tools and manipulators which are essential for both ongoing operations and remote repair/replacement and maintenance, which in turn exacerbated the reliability and availability issues with those components.

Some of the key lessons learned from this experience include:

1. The feed specifications, the process operating envelope and on-plant Quality Control and Quality Assurance necessary to demonstrate that the vitrified product meets specification must be developed, defined and optimized together.
2. Active glass sampling or non-destructive testing of the vitrified product is not necessary.
3. Operation within the process operating envelope is essential and therefore adequate margins which reflect the inherent accuracy of the measuring equipment must be incorporated.
4. Life expectancy cannot be tested during start-up, and may not be revealed in inactive testing. The FSIF melter was not an exact replica of the French-sourced AVH elliptical melter. However, the French development program, which did have a full scale AVH melter, did not reveal the corrosion problem that shortened early melter life.
5. All nuclear thermal treatment plants are challenging to operate. When the waste is very highly radioactive then the challenges are increased by orders of magnitude. The fundamentals must be demonstrated with sound R&D, thorough facility commissioning is paramount and plant performance can be improved incrementally with ongoing engineering and technical support.
6. Borosilicate glass can incorporate a wide variety of HLW compositions, has good chemical, mechanical, and thermal properties, and yields a consistent product. It was found to be optimum for the Sellafield wastes both in terms of product quality and on-plant processing. The R&D showed that

other base glass formulations could be tailored to deal with specific wastes.

Vitrification Plant Improvements

Recently British Nuclear Group entered into a new agreement with Areva-Cogema, the original suppliers of the vitrification cell equipment, to receive details of improvements implemented on their R7/T7 facilities in their La Hague reprocessing facility in northern France, and to develop further improvements for the Sellafield plants. In the 15 years that British Nuclear Group has operated the vitrification plants many hundreds of changes and improvements have been made. Some specific key improvements are listed below:

1. Initially melter life at 1200 hours average was considerably less than the design basis 2000 hours. Corrosion rates were significantly greater than expected. Thicker walled melters together with a different fin designs now enable a melter life of >3000 hours to be achieved. Designs are now being tested without internal fins.
2. An improved control system was developed for the melter power supply which enabled better temperature control. On lines 2&3 the 50Hz heating system has been replaced with a lower frequency system, enabling smoother temperature control.
3. The design of the power supply connectors to the melter have been significantly changed, improving both remotability and eliminating a susceptibility to a single point failure.
4. Calciner improvements have included more robust heater half shell design, improved roller bearings for longer life and consideration is being given to a more robust rabble bar design. An improved lower end fitting design was back fitted shortly after hot operations commenced on Line 1.
5. Considerable difficulty has been encountered due to the high solids loading to the primary off-gas. The frequency of blockages has been much greater than expected and has required much more frequent operational wash-outs than anticipated. Maintaining optimum flowsheet conditions has been difficult given the requirement to use the off-gas pipe wash much more frequently. Optimum dust scrubber conditions require high temperature and acidity. The dust scrubber recycle constant volume feeder (pump) has also been vulnerable to blockages and mechanical means have had to be deployed to remove them on a number of occasions.
6. When WVP Line 3 was built, many improvements were incorporated to facilitate remote maintenance, removal and replacement of the in-cell equipment. The principal changes were inclusion of two cranes to service the vitrification and breakdown cells, with hoist parks at each end of the line. Main crane decontamination facilities were included, together with improved supplementary tools and cranes to perform the crane maintenance. Line 3 has windows and master-slave manipulators on both side of the main process cell.
7. The most significant improvement in the container handling area is the provision of bead blasting to achieve satisfactory decontamination of the product containers. The original intent was to use high pressure water jets, but extensive repeat cycles were required. Improvements have also been made to the bellows seal device and its extract system which shrouds the product container during filling, thus reducing the amount of contamination coming into contact with the hot product container.

Full Scale Inactive Testing

In order to secure further process improvement and understanding, British Nuclear Group decided to re-commission the Full Scale Inactive Testing Facility (FSIF). The original facility had replicated the original WVP plant design, which was based upon the French Vitrification Plant located at the Marcoule site in southeastern France. The new FSIF accurately replicates the current WVP which, as described above, is based upon the La Hague Vitrification plant. This is perhaps the most significant lesson learned! The purpose of the current FSIF program is:

- To identify and validate process and equipment changes and improvements that will increase the throughput of the three Sellafield vitrification lines,
- To validate plant operating conditions so as to allow the plant to operate at increased throughput,

- To examine the limitations of the operating envelope, particularly with a view to increasing waste oxide incorporation above 25%,
- To demonstrate that the vitrified product quality will meet the specification at the increased throughputs/incorporation levels.

CONCLUSIONS

1. Considerable quantities of high level liquid waste have been safely and successfully stored, treated, and prepared for final disposal at Sellafield in the United Kingdom. A key element in this successful waste management strategy has been the incorporation of sufficient redundancy and diversity in processing capacity, operational and protective systems.
2. Considerable investment has been made in R&D to support the HLW program in the UK. Particular emphasis has been given over many the years to proving designs on full-scale test rigs and then testing key equipment prior to installation under normal and fault conditions. In order to accurately replicate fluid dynamic conditions, particular emphasis has been given to accurately physically replicating complete systems for pre-installation testing. This has proved essential for emergency emptying, agitation, sampling, gas clean-up and glass making equipment, sometimes revealing deficiencies in suppliers’ tests but more importantly often pointing the way to more optimum solutions and applications.
3. Numerous operational problems have been encountered during the 50+ years of HLLW operations. On occasions they have halted production and required equipment redesign, provision of additional equipment and protective measures, revision to operating procedures or additional or special measures to be taken. Although some of these problems have actually been classified as significant events by the Sellafield Operators, none have resulted in significant harm to any operator, the public or the environment.
4. The lessons learned are huge, many of which are directly relevant to the HLW programs in the US. The “owner-operator” culture has ensured excellent feedback between operators and designers and enabled maximum benefit to be taken from the extensive operational experience, some of which has been painfully gathered.

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