

Application of Modular Design in Abatement of Fuel Element Debris (FED) Liquid Waste Stream - 16635

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ABSTRACT

This paper provides a description of the processes applied within the Aqueous Discharge Abatement Plant (ADAP) at Bradwell nuclear power plant in the UK. This incorporates the Fuel Element Debris (FED) abatement plant and the Modular Active Effluent Treatment Plant (MAETP). These plants are housed in adjacent, but separate, structures and are interlinked and share common services and control systems. The FED abatement plant, which has a shorter mission than MAETP, can be removed for decommissioning or redeployment without any impact to MAETP operation.

ADAP treats aqueous waste streams generated during the ongoing decontamination and decommissioning (D&D) operations for release from the Bradwell Power Station. The plant is designed to receive and treat two major classes of liquids:

1. Slightly contaminated water from various D&D sources and collected rainwater from the power station, referred to as Aqueous Effluent (AE).
2. Dissolved fuel element debris solution. This comprises of contaminated magnesium alloy (generated during the stripping of external structures from spent Magnox fuel elements prior to shipment for reprocessing) which has been dissolved in nitric acid.

All received wastes are treated to remove heavy metals and radioactive constituents prior to isolation, sampling, verification, and ultimately release to the Blackwater Estuary. The treatment consists of multiple processes including precipitation, filtration, adsorption and ion exchange carefully designed to remove targeted radionuclides and heavy metals.

INTRODUCTION

Bradwell is a former Magnox nuclear power station which is currently undergoing D&D activities. A major source of radioactive waste on the site was the fuel element debris which had been generated during the lifetime of the plant until defueling operations were completed in 2006. Approximately 575 m³ of fuel element debris was stored in vaults and a Best Practical Environmental Option (BPEO) study indicated that dissolution of this solid waste in dilute nitric acid followed by treatment of the effluent was preferable as opposed to direct encapsulation [1]. Two types of FED were found to predominate: Zr55 Magnox (magnesium alloy containing approximately 0.55% zirconium) and Al80 Magnox (magnesium alloy containing approximately 0.80% aluminium). As a consequence, both the procedure developed to dissolve the FED and the waste treatment process needed to be able to accommodate both types of alloy.

In addition to the FED abatement plant, there was also a need to treat effluent resulting from D&D operations and rainwater that could potentially be contaminated. This was the design basis for the MAETP plant. MAETP took over the discharge routes for the original Active Effluent Treatment Plant (AETP) and the associated Pond Water Treatment Plant (PWTP) which historically provided cesium abatement for the site as well as maintaining pond water quality. (This latter plant was isolated and de-planted as part of the site D&D program.) The new MAETP plant is a replacement for the existing Active Effluent Treatment Plant (AETP) and provides a discharge route via the Final Monitoring and Delay Tank (FMDT) for waste discharge to the Blackwater Estuary for the Bradwell site to support current operations and future care and maintenance (C&M) operations. The MAETP is designed to ensure radionuclides are reduced to acceptable levels as defined by the regulatory authorities.

Feed Specifications

The effluent treatment processes described were developed based upon the feed characteristics and treatment requirements supplied by Magnox to EnergySolutions [2]. These were as follows:

FED Abatement Plant

The FED abatement plant must isolate and places into safe containment heavy metals and radionuclides supplied to the plant by Bradwell site's Magnox Fuel Element Debris Dissolution (FEDD) plant. The FED abatement plant feed stream comprises an inorganic solution in low molarity nitric acid (HNO_3) where the primary dissolved ionic constituents are magnesium ($25 - 50 \text{ g l}^{-1}$) and nitrate with heavy metals and radionuclides present in the ranges specified by elemental analysis. The process was designed to isolate heavy metals within a filter cake whilst leaving the magnesium in a neutralised solution for discharge into the Blackwater Estuary. The radionuclide inventory in the waste streams is concentrated in a filter cake and spent ion exchange media and granulated activated carbon (GAC), contained and stored within MOSAIK[®] ductile cast iron containers as proscribed by the customer. The waste forms must also be classified as Intermediate Level Waste (ILW).

The influent stream physiochemical characteristics the plant must be able to tolerate are outlined below.

- Particulate size range: less than $1 \mu\text{m}$ up to 3 mm
- Total oil in feed: trace levels only
- pH range: $0.5 - 2$
- Temperature: $<50 \text{ }^\circ\text{C}$
- Plant receipt flow rate $30 \text{ m}^3\text{hr}^{-1}$ (maximum)

Post treatment the radioactive aqueous discharges from the plant must be in compliance with the permit issued to Magnox by the Environment Agency (EA) under the Environmental Permitting Regulations 2010 (EPR 2010) [3].

MAETP Plant

The MAETP operates on the basis that any or all of the unit operations can be bypassed by manual intervention based on a decision by the plant manager from

knowledge of the waste streams as they arrive. This is to remove the burden of unnecessarily operating all of the facility structures, systems and/or components together, when simple filtration (e.g. for rainwater washings) may be all that is required to generate an effluent suitable for discharge.

The MAETP is used to deal with a wide range of influent streams from the Bradwell site from proposed future operations and as defined in a BPM study carried out in 2010 [4]. In radiological terms, the feed streams to the MAETP will be dominated by Cs-137 with lesser activity from Fe-55, Sr-90 and Pu-241. It is also possible that minor arisings containing Co-60, Ni-63 and others will be an occasional challenge that will need to be addressed by the plant. The expected radionuclides are removed by a combination of the filtration steps and the ion exchange system.

A summary of the waste characteristics the MAETP will be required to treat is:

- Activity range: 0–2 GBq/m³, (~90% of waste will be <0.1 GBq/m³)
- Particulate size range: less than 1 µm up to 3 mm
- Average particulate concentration: 100 ppm
- Maximum particulate concentration: 2000 ppm (for 99% of influent)
- Total oil in water: trace levels
- pH range: 6 – 12
- Conductivity range: 30 – 3000 µS/cm
- Temperature: <35 °C
- Plant throughput: 2–8 m³hr⁻¹. In the cases where the high throughput of 8 m³hr⁻¹ is expected, these are likely to be low activity liquids requiring filtration only. Higher activity fluids are treated at lower throughputs and the minimum flow for design purpose is 2 m³hr⁻¹.

The plant feed envelopes described above are taken from the technical specification provided by the Bradwell site [5] and the plants are designed to operate within these parameters. All metallic components in contact with process solutions are stainless steels (e.g. 316L stainless steel) which is capable of withstanding a wide range of chemical feed streams.

FED ABATEMENT PLANT PROCESS DESCRIPTION

The FED abatement plant operates on a batch basis, and receives acidic solutions from the Bradwell FED dissolution facility that contain relatively high levels of activity in solution. The activity can vary considerably from batch to batch and will depend upon the age and type of FED that has been dissolved. Three gamma emitting isotopes – Co-60, Cs-137 and Am-241 – are typically reported with individual activities typically ranging from 0.2 to 4 MBq/m³ or greater. The heavy metals in the solution are precipitated out by a neutralisation reaction and are then flocculated and allowed to settle as slurry which is isolated and dewatered to provide retrievable ILW and directed to a MOSAIK®. The supernatant liquor is filtered and passed through GAC and ion exchange media in order to ensure that the site discharge limits are met.

Feed Receipt

Effluent from the FED dissolution is received into ADAP via one of two 5.5 m³ buffer storage tanks. The tanks are identical, and both fitted with vertically oriented (cantilevered) long shaft centrifugal transfer pumps capable of recirculating the waste within the tanks, and thereby mixing the contents to keep any solids in suspension. These buffer storage tanks are capable of receiving two daily batches of approximately 5.3 m³ of feed from the dissolution plant. This feed solution is approximately 0.2 m nitric acid with a pH of approximately 1. All process vessels, including the buffer storage tanks, are fabricated from stainless steel.

Chemical Dosing

A 3 m³ batch of the feed solution in the buffer tanks is transferred into one of two 4 m³ reaction tanks. The reaction tanks are each fitted with vertically oriented (cantilevered) long shaft centrifugal transfer pumps and variable speed paddle mixers. The tanks are identical and both have a bottom drain. The pH is then carefully adjusted to between 7.2 and 7.8 using 4M sodium hydroxide solution. (A nitric acid feed is also available in case of pH overshoots.) This neutralization results in the precipitation of the bulk of the dissolved heavy metals as hydroxides while maintaining the magnesium in solution. It was noted during laboratory trials that cesium, technetium and a portion of the ruthenium in the feed material remained in the liquid phase. This metal hydroxide precipitate consists of very fine particulate so a flocculation polymer is added to the neutralized solution to coagulate the fine particles into sufficiently large particulates that gravity settling will occur. From the trials undertaken and subsequent plant data, the settled sludge is approximately 5-7% by volume of the solution, comprising 0.7 – 0.9 wt% solids. After the particulates have settled, the sludge layer is removed from the reaction tank via the bottom drain using an online turbidity meter to determine when all the sludge been drained from the tank.

The solids are pumped using low shear pumps from the reaction tanks to a small 0.75 m³ sludge holding tank, which acts as a buffer, ahead of the upstream dewatering process. After the settled solids are drained from the reaction tank, the remaining liquid is transferred to the microfilter feed tank.

Microfiltration

The supernatant liquor from the reaction tanks (and the permeate from the upstream filter press) is fed in batches to the 1.52 m³ microfilter (MF) feed tank. This is a metered process controlled by high and low level switches to ensure the MF feed tank is continually filled until the batch tank has been emptied. It also acts to prevent passage of air through the filter using the low level switch. The liquid is then passed through the microfiltration filtration unit. This ensures that the liquid is free of fines which could potential foul the downstream ion exchange media. The microfiltration unit is equipped with a backwash capability in the event it becomes blinded with fines. Inactive testing and plant operation showed excellent solid-liquid separation and rapid filtration of supernate with no filter blinding. Any backwash solutions from the MF unit generated during cleaning operations are redirected either to the FED Reaction or the FED buffer storage tanks.

Ion Exchange

Ion exchange is used as polishing activity since the bulk of the radionuclides of concern (cesium being the exception) are removed during the precipitation stage. These radionuclides of concern are Co-60, Fe-55, Ni-63, Cs-137, Cs-134, Pu-241, Am-241 (and other actinides). Sr-90 will be removed to a small extent by co-precipitation with the metal hydroxides but is not considered a radionuclide of concern as it contributes <2% of the discharge authorization. Ruthenium is not well removed but is a very minor contributor to discharges and is therefore not slated for ion exchange abatement. Critical nuclides for ion exchange abatement are Cs-134 and Cs-137, with Co-60 being a nuclide that may require polishing.

Three media beds are used to polish the liquor. The first is a Granulated Activated Carbon (GAC) column designed to remove any remaining colloidal solids, organics, organonucleide complexes and radioactive iodine. The second column is Cs-Treat media, a granulated KCo hexacyanoferrate. Cs-Treat media is intended to selectively remove cesium from the FED effluent stream and is non-elutable. Spent Cs-Treat media is discharged and replaced with fresh media as it becomes exhausted. Due to the high cost of the Cs-treat, it is essential that this media be used efficiently. It should be noted that the capacity of the Cs-Treat media greatly exceeds the cesium present in the FED vaults and the Cs-Treat will be replaced in accordance with the allowable inventory for Cs-137 in an IP2 Mosaik. The final column in the ion exchange train contains the Co-Treat media. Co-Treat media is intended to selectively remove cobalt from the FED effluent stream. Like Cs-Treat, Co-Treat media is non-elutable and therefore once the media is fully loaded to the limits of the IP2 Mosaik, the media is discharged and replaced. The flow rate through the IX system is designed to be 100 liters per minute.

Effluent Sentencing

After passage through the ion exchange columns, the effluent then goes through a final 50 micron bag/cartridge filter to capture any ion exchange media fines or other solids and then to an unshielded sentencing tank. A gamma monitor is fitted within the confines of the container (secondary containment) and within biological shielding, on the transfer line to the 25m³ Effluent Sentencing Tank. This treated effluent is isolated, sampled, and verified to meet release criteria prior to discharge to the Final Monitoring Delay Tanks (FMDT). Non-compliant effluent can be transferred back to the FED buffer storage tanks, in the unlikely event reprocessing is necessary. Sampling is performed to ensure the activity levels are within prescribed levels prior for onward transfer and discharge. In addition, sample analysis is used to monitor the performance of the upstream ion exchange media.

Filter Press / MOSAIK®

The use of a filter press to concentrate the sludge was based upon the success of this technology in the non-nuclear industry where analogous metal hydroxide sludges are routinely dewatered to generate a moist cake. By conventional industry standards the sludge load on the ADAP filter press is low and well within the typical process envelope for this technology. In the ADAP process, a 0.75m³ sludge holding tank, duty and stand-by positive displacement pumps, and the filter press are supplied as a complete packaged system. The press is a commercially available unit, with a proven industrial track record. Certain features of the unit are

enhanced, to facilitate 'safe' maintenance, and to assure confinement and containment are maintained at all times.

The filter press receives a predetermined volume from the sludge holding tank. The device relies on the pump pressure to compact the feed onto the plates and the integral membrane filters. Once the waste is dewatered, the resulting 'cake' is displaced by opening the plates within the press allowing the product to drop (by gravity) into the MOSAIK® directly beneath the discharge chute fitted to the press. The low turbidity permeate from the press is sent to the microfiltration feed tank and any turbid water resulting from filter press activities can be redirected back to the buffer or reaction tanks. The MOSAIK® is transported within the plant on a bogie system on rails to allow it to be repeatedly positioned within the facility.

MAETP PROCESS DESCRIPTION

The MAETP receives wastes from a range of sources on the Bradwell site including rainwater, shower drains, and legacy sources across the site, plus arisings from waste retrieval and decommissioning operations. The MAETP treats these wastes according to the characteristics of the influent streams and each of the individual treatment units within the plant has a bypass system, to divert the process feed to one, or each of the 3 unit operations. Decisions on the treatment options are made by suitably qualified operating plant personnel based on known characteristics of the upstream process feeds. This minimises the maintenance burden on the system as approximately 90% of the projected influent is expected to be rainwater which may only require filtration allowing the ion exchange beds and ultrafiltration system to be held in standby.

Receipt and pH Adjustment

The AE feed is initially passed through a 500 µm pre-filter screen for removal of coarse particulate material prior to receipt into one of three 5.5 m³ buffer storage tanks. This coarse screen is designed as a removable cartridge and will be transportable to a suitable receipt facility on site for disposal.

The three AE buffer storage tanks allow pH adjustment of the received AE stream with either nitric acid or sodium hydroxide to reach a target pH of 7.5. Each of the three buffer storage tanks are identical, and are fitted with vertically oriented (cantilevered) long shaft centrifugal transfer pumps configured to either transfer waste water from the tanks or to mix the waste water in the tank to keep any solids suspended in the waste water. Following buffer storage, all piping in the remainder of the AE treatment is configured to allow unit operations to be utilized or by-passed as needed to meet the release criteria.

Microfiltration

The feed from the buffer storage tanks is first passed into a 0.75 m³ microfiltration system feed tank. The microfilter consists of a backwashable cartridge filter designed to remove particulates ≥ 5 µm. In dead-head cartridge filtration, all of the fluid flow goes through the filter media. The control for the micro filtration unit operation relies on the measurement of the pressure drop across the filter. When the pressure drop reaches a set point, flow through the filter is stopped, and the filter is backwashed to remove the accumulated solids. The backwash stream containing the solids is connected to a 0.75 m³ backwash settling tank. After

backwashing the filter is returned to service. Over time, it is anticipated that backwashing will not return the filter to the initial flow/pressure condition due to solids build-up in the filter cartridge that cannot be removed via backwashing. After backwashing becomes no longer effective at restoring flow/pressure drop, the filter cartridge will be cleaned or replaced.

Ultrafiltration

The third unit operation in AE treatment is ultrafiltration. Ultrafiltration consists of cross flow filtration to remove particulates $\geq 0.1 \mu\text{m}$. Unlike dead-end filtration, in cross flow filtration the liquid passes through tubes constructed of filter media. Pressure drives the liquid through the porous media, and the cross-flow through the tubes prevents build-up of the filter cake on the filter media. Cross flow filtration includes a feed tank and pump. A flow rate of waste approximately 10 times higher than the desired filtrate flow rate is passed through the filter tubes. Approximately 90% of the flow through the tubes is returned to the filter feed tank and the remaining 10% passes through the filters as cleaned filtrate liquid.

Over time, solids may plug some of the pores in the filter media. When plugging occurs, cross-flow filters are designed to be back-washed by running permeate in the reverse direction through the membrane to remove surface fouling. This is an automated process controlled by differential pressure (DP) and performed while the filter is on-line. In addition, chemical cleaning can be performed as a manual process when the filter is off-line. This is achieved using an integral Clean-in-Place (CIP) packaged system, whereby wash solution is delivered through the (CIP) tank. The tank's cleaning solution is replenished when required. In backwashing, the filtrate is pushed through the filter media in the opposite direction from the normal filtrate flow. Over time the solids content in the liquid in the filter feed tank will increase and the filtrate flow through the filter will decrease. When the filtrate flow decreases because of the build-up of solids in the filter feed to the point where unit operations following the ultra filtration cannot be supported, the filter feed tank is drained to the settling tank. Backwashings are delivered to the 0.75 m^3 backwash settling tank. The target filtrate flow rate is approximately 100 l/min and the pressure drop through the filter system is approximately 340 kPa. The ultrafilter feed pump, and a booster pump on the filtrate line provides filtrate delivery to the downstream GAC and ion exchange columns.

The UF is designed to operate in both continual duty and batch duty. In continual duty the feed is concentrated in the system and a slipstream of concentrate is bled off continually. In batch mode the feed is continually concentrated by recycle to the Permeate Feed Tank (PFT).

Solids Processing

The solids flushed from the microfilter and ultrafilter systems are passed in batches to the backwash settling tank, where they are allowed to settle in a conical bottom tank. Spent ion exchange media and GAC will also be delivered to this tank. This system provides the best method of isolating solids without creating large volumes of secondary liquid wastes. Any solids collected can be routed to a 200 litre drum export directly underneath for disposal elsewhere on site, and remaining liquids (supernatant) is decanted intermittently back to the buffer feed tanks.

Ion Exchange

The liquor from the ultrafilter is passed through three beds of media to give the waste a final polish prior to sending to monitoring and discharge tanks. The beds in sequence are granular activated carbon (GAC), cation exchange media and a mixed ion exchange bed. This was later changed to GAC followed by two mixed bed ion exchange beds

The GAC bed eliminates any fines that pass the ultrafilter (e.g. colloidal Ni and Co) and also captures any organic materials in the liquor (e.g. trace oils). The GAC bed also acts to protect the ion exchange resins downstream from blinding.

The two ion exchange vessels remove the remaining activity and ions from the solution and serve to ensure the aqueous phase at this stage meets the requirements of the Environment Agency for discharge to the Blackwater Estuary. At this stage, a DF of 500 or better is achieved for most isotopes via the ion exchange processes. These vessels are intermittently monitored for build up of activity and recharged with fresh resin as the charge becomes spent.

The selection of media is based upon the influent specification and can be changed to introduce specificity for radionuclides of concern based upon feed stream inventory of radionuclides and counter ions. The system is not intended to operate with multiple waste streams from all Magnox operational feeds using a single ion exchange media. It is a facet of the functionality of the system that ion exchange media can be replaced in matter of an hour on a campaign basis.

Identical to the FED ion exchange and GAC columns, each vessel in the MAETP ion exchange system features a Johnson screen on the outlet to capture ion exchange media fines and so protect the downstream system from contamination. Local radiation instruments at the ion exchange vessels will indicate the build-up of radionuclides in the resins and will be used by operators to determine when to replace the spent media with a new charge of resin.

AE Effluent Sentencing

The treated AE effluent is passed to a sentencing tank where it is isolated and tested to verify it meets discharge criteria before being passed through to the FMDT tanks. This process is similar to that described previously for the treated FED liquor.

AE Solid Waste Handling

Backwashed solids, sluiced GAC and ion exchange media, are delivered to the 0.75m³ backwash settling tank. Supernatant from this tank is pumped back to the AE buffer storage tanks for reprocessing, and the settled solids are transferred into a 200 liter Drum via a transfer interface. Level indication within the settling tank will ensure the contents of the tank do not exceed 200 liters.

Common Systems to FED and MAETP

1) Chemical Feed Tanks

The chemical feed tanks are primarily used for the dosing of the FED process tanks, hence their proximity to them in the facility. All reagents are delivered through a

set of simple (diaphragm operated) metering pumps. There are pipelines leading to the MAETP buffer tanks. This is to allow adjustment of pH to ensure feed stream uniformity entering the process. The pH control system maximises precipitation efficiency and ensure optimal performance of the ion exchange beds as required.

2) HVAC

ADAP has a HEPA filter system attached to each process vessel and to each shipping container where it is routed to the ventilation outlet and operates at a minimum of five full air volume changes per hour. The influent air is conditioned if and as necessary to prevent condensation throughout the plant. At this time there is no requirement for a scrubber system as the process chemistry within the plant is expected to preclude the production of acidic gases.

3) System Flushes

Consideration is given to flush water systems and wash downs as appropriate to facilitate the reduction of activity prior to manual intervention for maintenance.

A photograph of the completed ADAP plant, comprising both the FED and MAETP units, is shown below in Figure 1.



Figure 1. ADAP plant at Bradwell power plant, UK

PLANT PERFORMANCE

Both the FED and MAETP plants have met design expectations in terms of isotope and heavy metal removal. The discharge limits for a number of heavy metals are shown below in Table 1. These levels are the maximum allowable concentrations in the FMDT tank and take account the fact that the discharge will be mixed with seawater during discharge, further diluting the concentrations.

| Element | Cr | Fe | Ni | Cu | Zn | Cd | Pb | Hg |
|-------------------------|-------|-----------|-------|--------|---------|----|------|-----|
| Limit / $\mu\text{g/l}$ | 4,563 | 7,605,000 | 3,000 | 38,025 | 304,200 | 30 | 1080 | 7.5 |

Table 1. Maximum allowable metal concentrations in the FMDT tank

As can be seen in Table 1, the limits for some metals are very challenging but the plant has consistently produced effluent well below these limits.

The gamma emitters Co-60, Am-241 and Cs-137 have routinely been reduced to below the levels of detection on both plants (typically 0.05 MBq/m³ or lower), exceeding the desired removal rates.

CONCLUSIONS

Both the FED and MAETP components of ADAP have proved successful in treating the radioactive waste streams generated by ongoing D&D operations at Bradwell nuclear power plant. The data from the FED plant has demonstrated that the liquor generated from the dissolution of Magnox fuel debris can be effectively treated using a combination of precipitation/filtration and ion exchange polishing to generate a final effluent suitable for environmental discharge. Both heavy metals and radioisotopes have been removed to well below discharge specifications allowing their safe, controlled release to the Blackwater Estuary. This has resulted in a substantial decrease in the level of ILW requiring disposal. Once the fuel element debris dissolution campaign is completed, the modular nature of the plant means that the FED abatement part of ADAP can be decommissioned without impacting throughput of the MAETP plant which will be continue to be required to support plant operations.

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