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# AERIAL AND LIQUID EFFLUENT TREATMENT IN BNFL'S THERMAL OXIDE REPROCESSING PLANT (THORP)

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## Abstract

British Nuclear Fuels plc (BNFL) completed construction of its Thermal Oxide Reprocessing Plant (THORP) at Sellafield in 1992, at a cost of £1,850M. After Government and Regulatory approval, active commissioning was initiated in January 1994. Since then, the whole of the plant has been progressively commissioned and moved towards full operational status.

From the outset, the need to protect the workforce, the public and the environment in general from the plant's discharges was clearly recognised. The design intent was to limit radiation exposure of members of the general public to " As Low as Reasonably Practicable" (ALARP). Furthermore no member of the most highly exposed (critical) group should receive an annual dose exceeding 50 microsieverts from either the aerial or marine discharge routes.

This paper describes how the design intent has been met, concentrating mainly on aerial discharges. It describes the sub-division of the plant's ventilation system into a number of separate systems, according to the volume and source of the arising and the complexity of the treatment process. The dissolver off-gas, central off-gas, cell and building ventilation systems are described, together with the development programme which was undertaken to address the more demanding aspects of the performance specification. This ranged from small-scale experiments with irradiated fuel to inactive pilot plant trials and full-scale plant measurements. In addition wind tunnel tests were employed to assist dispersion modelling of the gases as they are discharged from the THORP stack. All the resulting information was then used, with the aid of mathematical models, in the design of an off-gas treatment system which could achieve the overall goal.

As far as liquid discharges are concerned, the paper describes the overall philosophy behind the THORP flowsheets which allows the environmental impact target to be met. The treatment of some waste streams is undertaken in dedicated equipment within the THORP building. Other streams are mixed with effluents from elsewhere on the site and treated in existing site facilities. After treatment and sentencing the effluents containing low levels of radioactivity are discharged down a 3 km pipeline into the Irish Sea.

Taken together, the aerial and liquid discharges from THORP are predicted to fall well within the original design intent of 50μSv. Aerial discharges are predicted to give a maximum critical group dose of 22μSv and marine discharges will give a dose of 36μSv to a separate critical group. Moreover the discharges are also predicted to fall comfortably within the limits on individual radionuclides which have been imposed as part of the Government's Aerial and Liquid Discharge Authorisations.

The detailed attention given to the development and design phases of the effluent treatment system should ensure that these predictions are mirrored by actual performance as the plant becomes fully operational.

## INTRODUCTION

### Brief History

THORP is the third reprocessing plant to be constructed on BNFL's Sellafield site in Cumbria, UK. The plant is designed to reprocess irradiated fuel from Advanced Gas-Cooled Reactors (AGR) in the UK and from Light Water Reactors (LWR) in Europe and Japan: virtually all of its first 10 years operation are committed with contracts for 7000t(U).

Conceptual design of the plant started in 1974, supported by construction and operation of a number of major development facilities during the later 1970's and early 1980's. Design of the plant was delayed by the Government's decision to hold a public inquiry (the Windscale Inquiry) in 1977. Following the successful outcome of the inquiry, design recommenced in 1983, leading to the start of civil construction in 1984 and eventual handover of the completed plant in 1992. Plant commissioning then proceeded but the progression to active commissioning, planned for December 1992, was delayed by a protracted public consultation exercise to satisfy recent environmental legislation. Government and Regulatory approval to proceed was finally given, leading to the commencement of active commissioning on 17 January 1994.

### Design Intent

All BNFL's operations on the Sellafield site are strictly controlled by Government Authorising Bodies. For the control of discharges to the environment, the relevant bodies are Her Majesty's Inspectorate of Pollution (HMIP) and the Ministry of Agriculture, Fisheries and Food (MAFF). Certificates of Authorisation issued by the Departments impose strict conditions on the discharge and disposal of radioactive waste. They also require BNFL to carry out detailed environmental radioactivity monitoring programmes (in addition to their own independent measurements) and to use "Best Practicable Means" to further limit the radioactivity in the waste discharged.

BNFL's strategy for the management of aerial and marine radioactive effluents arising from THORP operations were first formally stated at the Windscale Inquiry in 1977. In summary the design intent was that:

(a) radiation exposure of members of the general public, arising from discharges from THORP and associated facilities, should be "As Low As is Reasonably Practicable" (ALARP);
(b) in any case, no member of the most highly exposed (critical) group should receive an annual committed effective dose equivalent (CEDE) in excess of 50μSv from either aerial or marine discharge routes.

In the context of exposure to the general public, it is important to consider both:

(a) individual risk, as measured by doses to members of the critical group, and
(b) societal risk, as measured by the collective dose to large populations over a long time period.

In addition to dose uptake constraints, HMIP has more recently set an Aerial Discharge Authorisation for THORP which limits the quantity of specific radionuclides which can be released from the THORP stack on a daily, quarterly or annual basis. The annual discharge limits for the most significant radionuclides are given in Table 1.

As far as liquid discharges are concerned, HMIP's Authorisation relates to the whole site rather than THORP specifically and is depicted in Table 4.

**Scope of Paper**

This paper describes how the design intent on aerial and liquid discharges has been met. Concentrating primarily on aerial discharges, it examines the different sources of off-gas arisings from the THORP process and describes how the different types of stream have been segregated for treatment prior to extraction and discharge from a common 125m stack. The development work associated with some of the key process equipment is described and key design features of the equipment are covered.

As far as liquid discharges are concerned, the paper describes the overall philosophy behind the THORP flowsheets which allows the environmental impact target to be met. The treatment of some waste streams is undertaken in dedicated equipment within the THORP building. Other streams are mixed with effluents from elsewhere on site and treated in a variety of existing or new site facilities, which are described in outline.

Finally, the paper compares the predicted aerial and liquid discharges from the plant with the latest Discharge Authorisations to illustrate how well the plant design is expected to meet its objectives.

**Table 1 - Comparison of Predicted THORP Aerial Discharges with Discharge Authorisation**

| Radionuclide | THORP Discharges (TBq/yr) | THORP Downstream Plant Discharges (TBq/yr) | THORP[(1)] Total Discharges (TBq/yr) | THORP Authorisation (TBq/yr) |
|---|---|---|---|---|
| H-3 | 21.6 | 0.0007 | 21.6 | 43 |
| C-14 | 0.434 | ~0 | 0.434 | 0.87 |
| Kr-85 | 369,000 | 6.61 | 369,007 | 470,000 |
| Sr-90 | 0.004 | 0.0036 | 0.0076 | 0.0078 |
| Ru-106 | 0.0355 | 0.0016 | 0.0371 | 0.05 |
| I-129 | 0.0218 | 0.0031 | 0.0249 | 0.044 |
| Cs-137 | 0.0055 | 0.005 | 0.0105 | 0.011 |
| Pu(Alpha) | 0.00027 | ~0 | 0.00027 | 0.0005 |
| Total Alpha | 0.00048 | ~0 | 0.00048 | 0.001 |
| Total Beta (β5) | 0.152 | 0.017 | 0.169 | 0.28 |

Critical Group dose [(1)] = 22μSv/yr
Target dose = 50μSv/yr

(1) Based on 1200t(U)/yr of reference fuel

## GASEOUS ARISINGS

THORP is an integrated reprocessing plant with all the major process steps being carried out in a single building (See Fig 1). Only where use could be made of waste and effluent treatment facilities serving existing process plants on the Sellafield site are certain operations performed outside the THORP building. The THORP process breaks down principally into a shear/leach "Head End" section with clarification of the dissolver solution, followed by chemical separation of the uranium, plutonium and fission products by solvent extraction in pulsed column contactors. The uranium and plutonium products are purified by further solvent extraction prior to conversion to solid forms suitable for storage or return to customers.

Within the plant as a whole, a variety of different off-gases arise.

### Dissolver Off-gas

Leaching of the sheared fuel in boiling nitric acid releases virtually all the volatile components of the irradiated fuel assemblies, principally I-129, C-14 and Kr-85. Also Nitrogen oxides (NOx) are generated by dissolution of the fuel. The dissolver off-gas (DOG) stream consists of the ventilation air from the shear machine and the off-gases from each of three dissolvers, amounting to a flow of about 400$m^3$/hour. The stream also contains very fine particles of fuel dust which are not captured in the dissolver and, in total, represents by far the biggest off-gas challenge in THORP.

### Central Off-gas

Apart from the dissolvers, most of the other vessels and process equipment within the Head End and Chemical Separation Plant are vented via a separate Central Off-gas (COG) system. This has to accept a large number of streams with widely differing flows and compositions.

The most environmentally significant component of the COG arisings is I-129. Although the vast majority is volatilised into the DOG system, any residual amount is assumed to be released from downstream vessels into the COG system. In addition, aerosols created in the chemical treatment and liquor transfer processes throughout the plant carry particles of non-volatile actinides and fission products into the ventilation stream. The nuclides that contribute significantly to the aerial activity levels are Ru-106, Pu, Sr-90 and Cs-137.

There are certain non-active arisings which have to be considered in the design of the COG system. The only one subject to regulatory discharge limits is NOx.

### Cell Ventilation

THORP is designed on the principle of cascading depressions between areas to provide barriers against spread of contamination. Cells and caves which contain the most highly active processes in the plant are therefore maintained under a depression with reference to the adjoining access areas. Generally inleakage at cell depression is adequate to provide the air flow required in a cell with little or no heat emitting equipment. In cells with large heat sources, additional air is provided to dissipate the heat by purpose-built engineered inlets comprising HEPA filters and control/fire dampers.

Air extracted from cells and caves is assumed to exhibit the same distribution of radionuclides (footprint) as is present in the process equipment in the cell. Arisings from mechanical operations of fuel shearing in head end caves have been assessed on empirical data from similar operations on other plants. In cells where operations are normally clean (washdown provisions are installed to clean-up spillages), the arisings have been based on potential re-suspension of liquor from pools on the bunded floors.

**Building Ventilation**

Building ventilation and heat systems provide a comfortable working environment in 'normal' and 'limited' access areas of the plant.

Normally accessible (unrestricted access) areas, despite having no definitive arisings (except under abnormal circumstances), are allocated an assumed mean airborne contamination which, if inhaled over a 2000 hour year, would result in 1% of the whole body dose target being received by inhalation.

Restricted access areas normally have very low arisings but have the potential for activity being present for short periods in abnormal circumstances. Such areas are allocated an assumed mean airborne contamination of ten times that allocated to the normal access areas. Under such conditions personnel would wear respiratory protection. In each instance the nuclides present are in proportion to the 'footprint' of the active material being processed.

**Total Ventilation System**

A schematic diagram showing the inter-relationship of the various components of the THORP ventilation system is given in Figure 2. It illustrates that main ventilation streams are kept separate until they enter the 125m stack from which they are discharged.

## FLOWSHEET AND EQUIPMENT DEVELOPMENT

**Dissolver Off-gas**

The prime task of the DOG system is to remove nitrogen oxides generated by the dissolution of the UO2 fuel, together with the major volatile radioactive species released as the fuel is dissolved. The DOG challenge is illustrated in Table 2, together with the flowsheeted Decontamination Factors (DFs) for each item of equipment.

**Table 2 - DOG System Performance**

| Radionuclide or species | Arising[1] (TBq/yr) | Flowsheet Decontamination Factor (DF) | | | | |
|---|---|---|---|---|---|---|
| | | Condenser | Acid Scrubber | Caustic Scrubber | Weak Acid Scrubber | HEPA |
| H-3 | 97.2 | 1 | 3 | 1.5 | 1 | 1 |
| C-14 | 28.9 | 1 | 1 | 70 | 1 | 1 |
| Kr-85 | 3.69E5 | 1 | 1 | 1 | 1 | 1 |
| Ru-106(gas) | 37.5* | | 20 | 100 | 1 | 1 |
| Ru-106 (solid) | 37.5* | | 1 | 1 | 1 | 10E4 |
| I-129 | 1.41 | 1 | 1.05 | 100 | 1 | 1 |
| NOx | 8.1E4 $m^3$ | 1.5 | 3 | 100 | 1 | 1 |
| Fuel Dust | 2.6E3 kg | 25 | 70 | 1.2 | 1.4 | 10E4 |

* post condenser (1) Based on 1200t(U)/yr of reference fuel

Recognising the need for highly efficient I-129 removal in THORP, the first requirement is to volatilise as much as possible at the dissolver stage. Once converted to organic forms by contact with solvent (TBP/OK) further down the process, iodine capture is much more difficult. Experimental work has demonstrated that the NOx evolved during dissolution of the fuel is effective in maintaining the I-129 in its volatile molecular form, which renders it easier to remove from the off-gas system.

Treatment of the DOG stream is performed principally by acid and caustic scrubbing in packed columns. The full train of DOG equipment is shown in Fig 3

The off-gases from each dissolver pass through their own dedicated reflux condenser and are then combined to be fed to the recombined acid column. Here, scrubbing with 6M nitric acid serves to remove the bulk of the oxides of nitrogen and two impingement plates at the base of the column assist removal of the entrained fuel dust.

A small amount of the iodine is also absorbed in the acidic scrubber liquor and this has to be removed before the liquor is fed back into the main process stream. This iodine desorption is achieved by heating a bleed of scrubber liquor and passing it through a small diameter packed column, counter current to a flow of air, which then joins the main off-gas flow carrying the desorbed iodine.

Following the recombined acid column is a large empty vessel, termed the "noxidiser", which provides a residence time of about two minutes in which the nitric oxide present is substantially oxidised to nitrogen dioxide by the oxygen in the air (which makes up the major proportion of the off-gas flow).

Increasing the proportion of nitrogen dioxide in this way improves the absorption of the nitrogen oxides in the caustic scrubber immediately downstream. The caustic scrubber will achieve a reduction in the nitrogen oxide concentration to below 1000ppm and a reduction in the I-129 and C-14 by factors of 100 and 70 respectively. Following the caustic scrubber the gas stream will be scrubbed with chilled recirculating water to remove entrained caustic soda droplets and dehumidify the gas. Finally the DOG stream undergoes two stages of HEPA filtration for final particulate removal before discharge to atmosphere.

**DOG Supporting Development**

In view of the dependence of the off-gas system upon aqueous scrubbing, the major facility in the development programme was a pilot scale rig - the dissolver off-gas pilot plant - to enable study of each of the scrubbing processes in the DOG system.

The rig, depicted in Fig 4, consisted of a small packed column, 100 mm diameter, with a packed height of 3m. Equipment was provided to generate a suitable mixture of oxides of nitrogen in air to which gaseous iodine could be added as appropriate. The scrub liquor could either be nitric acid or caustic soda solution and the liquor recirculated continuously through the column or treated in a single pass.

Sampling of the gases by means of heated sample lines enabled gas phase analysis by UV or IR spectrometers for nitrogen oxides and carbon dioxide respectively. Sampling for

iodine was achieved by selective absorption on heated beds of silver nitrate impregnated absorbers.

As originally conceived the pilot plant was to have handled iodine traced with I-131 in order to facilitate analysis to the low levels required. This increased significantly the complexity of the rig and would have made experimental work much more unwieldly. However, satisfactory development of ion selective electrode techniques sensitive to iodine concentrations as low as $1 \times 10^{-7}$ M removed the need for trace active work and considerably simplified the experimental programme. The outstanding analytical difficulty was the determination of inactive iodine on the silver nitrate absorbers used for sampling the gas streams but this was readily overcome by the use of neutron activation analysis.

Experimental investigation of acid and alkaline scrubbing of nitrogen oxides was supported by work on computer simulation of the processes in a package developed by BNFL. This package, NOXSIM, was thoroughly validated against the pilot plant experimental results and thus could be used with confidence in the design of the THORP off-gas columns.

Apart from the development work on the DOG pilot plant, other supplementary development was undertaken to address specific issues within DOG system. For example, experiments were performed to investigate iodine behaviour during dissolution of irradiated fuel and the subsequent treatment of dissolver off-gases. These investigations were part of the experimental work carried out on the THORP miniature pilot plant, a highly active 1:6500 scale pilot plant built primarily to study solvent extraction flowsheets. The dissolution of irradiated fuel served to show that the I-129 was present as molecular iodine and hence the simulation in the DOG pilot plant was valid.

Carbon-14 behaviour was less of a problem than I-129 since the bulk of it would be present as carbon dioxide and this would mix with the much larger quantities of $CO_2$ in air. There is abundant information on caustic scrubbing of $CO_2$ in air but a number of confirmatory experiments were necessary on the DOG pilot plant to ensure that the presence of 1 to 2 per cent NOx would not affect the $CO_2$ removal.

Not all the C-14 released from the dissolver is in the form of $CO_2$: a small amount is present as CO. Measurements performed during dissolution of irradiated fuel on the THORP miniature pilot plant confirmed literature information that the CO component was less than 1% and would not compromise the ability of the caustic scrubber to reduce C-14 levels by a factor of 70.

Measurement of $CO_2$ absorption has also proved important in the detection of potential maloperation. Experiments with the DOG pilot plant have shown that monitoring for increased $CO_2$ in the off-gases downstream of the scrubber provides adequate warning of approaching loss of alkalinity.

One issue raised well into the design phase of the THORP DOG system was a suggestion that troublesome quantities of volatile Ru-106 might penetrate into and even through the off-gas system. At first this question was explored by means of dissolution of irradiated fuel in a highly active cell and then in greater depth using trace active laboratory experiments. These two pieces of work were sufficient to indicate that only relatively small

amounts of volatile Ru-106 would be generated and that the off-gas system as designed would ensure adequate removal. Subsequent studies with irradiated fuel dissolution in the THORP miniature pilot plant have indicated that the removal efficiency in the THORP dissolver off-gas system will be ten times greater than initially estimated and thus the impact of volatile Ru-106 has proved to be of little significance.

Central Off-Gas

As already described, the COG system serves most of the vessels and equipment downstream of the dissolvers. The full train of COG equipment is shown in Fig 5. The off-gas arisings from different parts of the plant or from different types of equipment are combined into a series of " headers", which feed into the COG system at an appropriate point according to the type of decontamination required. The arising of key components in the COG feed is shown in Table 3, together with the flowsheeted DFs for each item of equipment.

**Table 3 - COG System Performance**

| Radionuclide or species | Arising[1] (TBq/yr) | Flowsheet Decontamination Factor (DF) | | | |
|---|---|---|---|---|---|
| | | Caustic Scrubber | ESP | Dehumidifier | HEPA |
| Sr-90 | 56.4 | 10 | 100 | 10 | 10E4 |
| Ru-106(gas) | 0.15 | 1 | 1 | 5 | 1 |
| Ru-106(solid) | 14.4 | 10 | 100 | 10 | 10E4 |
| I-129 | 0.009 | 2 | 1 | 1 | 1 |
| Cs-137 | 79.9 | 10 | 100 | 10 | 10E4 |
| Pu(alpha) | 6.19 | 10 | 100 | 10 | 10E4 |
| NOx | 84.9 $m^3$ | 43 | 1 | 1.6 | 1 |

[1] Based on 1200 t (U)/yr of reference fuel

First in the COG system is a 1.8m diameter caustic scrubber which serves the iodine header. This is primarily for iodine removal though it removes NOx and some particulate as well. A conservative Decontamination Factor (DF) of 2 has been assessed for iodine removal because the residual iodine from these downstream operations will be substantially organic. The scrubber performs a secondary function as an iodine trap in the event of accidental neutralisation of iodine-bearing streams in the Low Active Effluent Treatment Plant.

The next equipment in the train is a pair of 680 $m^3$/hr Electrostatic Precipitators (ESPs) whose purpose is the removal of entrained droplets and aerosols from the particulate header. These devices have proved to be very reliable and effective in other applications in highly active plant at Sellafield and only generate a low volume of liquid waste for treatment. Furthermore, by reducing the load on the HEPA filters at the end of the train, they reduce filter change frequencies and waste disposal costs. However care has to be taken to ensure that any solvent vapour in the feed stream is well below its lower flammability limit because the high voltage across the electrodes represents a potential ignition source.

The dehumidifier scrubber downstream of the ESPs is primarily aimed at removing moisture from the gas stream prior to HEPA filtration. This would cause deterioration of the

filter elements, leading to their eventual collapse and hence increased solid waste arisings. In order to prevent this the vent gas has to be cooled below its dew point, using a chiller unit to cool the dehumidifier scrubber liquor.

In addition, the dehumidifier acts as the primary disentrainment device for streams for which treatment in the ESPs is unnecessary. These include the low contamination vent header and the pulse header from the pulsed column drive legs in the highly active and plutonium purification cycles. With a large number of pulse systems operating independently the header could, theoretically, receive a very high flowrate if all the pulse systems discharged simultaneously. A Monte Carlo approach was used to predict the highest realistic flow likely to arise, thereby minimising the degree of over- design.

The final equipment in the train are banks of primary and secondary remote-change HEPA filters, which act as a final polishing step for particulates and as a back-up in the event of equipment maloperation upstream. Hot air is bled into the vent before the filters to ensure that the feed is well above its dew point.

The COG stream is finally discharged from a 125m stack, together with other THORP off-gases.

**COG Supporting Development**

In comparison with the DOG system, very little specific development work was necessary to support the COG system design. In relation to scrubber design, data generated from the DOG pilot plant were used, with the help of the NOXSIM computer model, to assist design of the COG unit.

One area which did require special attention was the characterisation of the aerosols generated by pulsing systems, agitation systems and various liquor transfer devices within the plant. Information on quantity and droplet size distribution was generated by development trials on replica equipment. This work showed that treatment of many of these streams in the ESPs was unnecessary; scrubbing in the dehumidifier gave sufficient decontamination.

Some development was performed on the ESP units during the design phase. Experience on earlier plants and collaboration with the manufacturer enabled a simpler and more compact design to be used on THORP without losing any efficiency. The simplification of the central electrode design within the collection tubes was demonstrated on a test rig.

**Glovebox Ventilation**

The finishing processes on the purified plutonium stream are largely conducted in glovebox containments. As gloveboxes with their high alpha inventories have to be protected against both overpressure and excessive depression under fault conditions, they are separately vented from the COG system.

The glovebox extracts are fitted with vortex amplifier (VXA) control devices operating under ambient pressure control to provide a controlled normal operating depression with small gas purge flows. VXAs and other fluidic devices are used extensively in THORP because their maintenance-free operation is invaluable in a highly radioactive environment. If a glovebox containment develops a leak or is breached the VXA device automatically opens to provide enhanced containment flow through the breach. These devices have proved highly

reliable on both plutonium glovebox and alpha laboratory analytical suites in existing Sellafield facilities.

The glovebox extracts are collected together and pulled through primary and secondary HEPA filters prior to discharge up the 125m stack in a dedicated flue.

**Cell and Cave Ventilation**

The plant is designed on the principle of segregation of ventilation streams of different activity levels to restrict the spread of contamination and minimise the amount of highly active waste materials.

Cell and cave air is extracted through fully shielded ducting to two active filter caves located in the centre building where the streams are filtered through 2-stages of HEPA filtration prior to discharge through the main stack. Several cells with minimal contamination potential have local manual-bag-change, circular filters, as do cells with predominantly alpha-bearing streams.

To prove the concept of the filter cave, prototype filter modules were constructed to minimise the pressure drop and improve the isolation damper maintenance problems. The grab assembly, which is essential for reliable filter change operations, underwent several revisions which were extensively tested to ensure the action was positive and reliable over a long period in a hazardous environment.

**Building Ventilation**

The largest proportion of THORP building is devoted to normally accessible work-areas and interconnecting corridors. The air required for ventilation purposes is supplied filtered at a constant temperature of 19°C in winter. Overall air change rates are inappropriate for use on large plants and would have resulted in an expensive and uneconomic installation. Each ventilated space within the THORP building has had its requirement for fresh air assessed on the need for heat, fume or humidity removal, occupation level and function. This approach has resulted in a smaller, more economic ventilation plant able to meet the large variations in individual room requirements.

To maintain the principles of containment, air is transferred from zones of low contamination potential to zones of higher contamination potential, thus minimising the spread of activity. Air extracted from the various zones is collected in separate dedicated ducting systems. In the case of the building air from beta-gamma areas, this is discharged unfiltered from short ducts terminating above the building parapet height. Air from alpha areas where there is normal access is passed through single stage HEPA filters before discharge from the main stack.

Air from limited access areas is extracted through single-stage HEPA filters in manually changed (bagged out) housings and discharged to atmosphere from the main stack. Limited access alpha areas are filtered through two stages of HEPA filters before stack discharge.

**Stack Dispersion Modelling**

The first detailed flowsheets identified a requirement for a stack capable of discharging 485,000 $m^3$/hour of effluent at a discharge height of between 80m and 120m based on a building height of 35m. A multi-flue connected to the main plant (centre building) was

proposed, the main ducts being contained within a bridge section which would be used for flow measurement and sampling/monitoring.

Wind tunnel tests were carried out to assess the performance of both stacks (80m and 120m) on a 1:200 scale site model, with varying windspeeds and directions and a range of discharge flue velocities. Due to the lack of sufficiently accurate gas concentration measurement equipment at this time, much of the work was done using high speed camera techniques with a neutral buoyancy smoke tracer in the flue discharge. The 120m high stack, employing a discharge velocity in excess of 30m/sec gave an effective height of 120m based on the visual appearance of the plume.

Further wind tunnel tests were carried out in 1988 to verify the performance of the stack based on final flowsheet values and using modern Flame Ionisation Detector systems which can detect levels an order of magnitude smaller than the earlier technology. In the interim period, the stack design had been simplified to one central flue of 3m diameter housing the major ventilation streams but incorporating COG and DOG streams at 108m level. The glovebox discharge joined the main flue at 108m level but continued inside the flue in its own discharge pipe to prevent back-pressure.

The later tests were used as a basis for quantitative effective height assessment, and demonstrated an effective height of only 85m for a 120m stack when buildings are present and in the most limiting weather conditions. Further tests with the model were able to show that an additional flue extension of 5m to give a height of 125m would produce an effective discharge height of 92m, which was sufficient to keep the critical group dose impact well below the target value and ensure the plume clears all site buildings. Thus the final design has a flue discharge height of 125m, a windshield roof level of 115m and a flue velocity of 34m/s.

## LIQUID ARISINGS

### Flowsheet Philosophy

Being the first new reprocessing plant at Sellafield for nearly 30 years, THORP was able to take advantage of modern thinking in flowsheet design, particularly with respect to minimisation of waste arisings. A number of clean technology concepts have been incorporated into the overall process in order to simplify waste treatment and reduce arisings.

A major improvement in the THORP flowsheet compared with earlier Magnox flowsheets is the move to salt-free reagents in the plutonium reduction stage. Traditionally, a ferrous salt such as ferrous sulphamate has been used to reduce plutonium to the non-extractable 3-valent state in order to effect a separation from uranium. This imposes severe restrictions on subsequent waste stream concentration because of the presence of iron salts. Process chemistry development for THORP identified U IV stabilised by hydrazine as the optimum reductant. Whilst not strictly salt-free, the added uranium is oxidised to U VI and follows the product stream with no impact on waste stream composition.

Similarly, the oxidising and reducing agents used in the downstream purification cycles are salt-free reagents, giving rise only to water and/or simple gases which have no deleterious effects on subsequent effluent treatment processes. Thus an even greater proportion of active waste isotopes can now be concentrated and subsequently vitrified. Apart from the raffinates from solvent washing, for which no satisfactory substitutes for sodium carbonate and sodium

hydroxide have yet been found, all the aqueous process effluents containing significant levels of radioactivity can be so treated.

The clean technology concept of effluent recycle has been used extensively in THORP. This is particularly true of the use of recycled nitric acid for many process feed streams. Acid recovery takes place in the salt-free evaporator system which processes all salt-free waste streams from the THORP process apart from the highly active raffinate from the first cycle (see below). The salt-free evaporator contains a fractionating column and produces three products, a concentrate containing virtually all of the activity, low acidity overheads which are discharged to sea and a low active recovered acid stream (6M $HNO_3$) which is recycled to the main plant. This recovered acid is used in virtually all acid feeds to THORP apart from those to the dissolver.

**Breakdown of Liquid Effluents**

There is a large number of liquid effluents arising at different points within the THORP process. The purpose of this paper is not to describe them all in detail but to give an overview of the types of streams and the methods of treatment adopted. THORP effluents can be categorised into three general types:

a) those which are normally suitable for sea discharge without activity reduction
b) those which are treated locally within THORP
c) those which are treated centrally by site facilities

**Discharges to the Segregated Effluent Treatment Plant (SETP)**

Over 30 streams from THORP are directed to SETP for sea discharge. Acidic and alkaline streams are segregated within THORP and transferred separately to SETP, where the acidic effluent is neutralised before mixing with alkaline effluent. Care has to be taken to avoid any risk of acidifying alkaline streams which contain Iodine-129, as this would probably cause volatilisation and release of this environmentally significant radionuclide. The combined effluents are then sentenced and discharged to sea.

Typical feeds to SETP include decontamination liquors from the THORP decontamination centre, condensates from the MA salt-free evaporator, excess recovered nitric acid, condensate from UN evaporation, COG caustic scrubber liquor and many others.

**Effluents treated locally in THORP**

Certain effluents require a dedicated treatment facility local to the source of arising in order to reduce the concentration of specific radionuclides before sea discharge. The first of these is the DOG caustic scrubber effluent, which requires removal of carbon-14 and plutonium present as fuel dust. As the carbon-14 is present as carbonate, along with absorbed atmospheric $CO_2$, the selected removal process uses precipitation as barium carbonate. This process was developed in the laboratory to establish optimum settling time, and washing parameters for the precipitate, together with an understanding of the behaviour of small particles of fuel dust in the process. The precipitate is encapsulated in cement as an Intermediate Level Waste and the low active supernate is sentenced and discharged to sea.

The second stream requiring local treatment is the Feed Pond purge. Feed Pond operations may result in suspension of some solids bearing cobalt-60 which would have a significant environmental impact. Therefore the purge is passed through a Funda filter for

solids removal before discharge to sea. It is noteworthy that pond water treatment for THORP is much simpler than for the earlier Magnox plants. Corrosion of stainless steel or Zircaloy cladding on oxide fuel is much less than for Magnox alloy, and the spread of any corrosion products is limited by intermediate containers. Hence ion exchange treatment of THORP pondwater is unnecessary.

The final category of effluents requiring local treatment in THORP are the Medium Active salt-free effluents. These are typically the raffinates from the product purification stages of the chemical separation plant, which are relatively high in activity but low in salts thanks to careful flowsheet design. These streams can be evaporated by a substantial factor (~50) without risk of crystallisation or precipitation in the concentrate. The overheads from the Medium Active salt-free evaporator are fed to a fractionating column for acid recovery, whilst the concentrate is fed to the site HA evaporator. This is a good example of clean technology by recycling streams within the overall process.

**Effluents Treated by Site Facilities**

Where suitable site facilities already exist, these are utilised for the treatment of appropriate THORP effluents. The facilities in question are:

(i) HA liquor evaporation and storage (HALES)
(ii) Evaporation of salt-bearing liquors (Salt Evaporator)
(iii) Enhanced Actinide Removal Plant (EARP)
(iii) Solvent Treatment Plant (STP)

The HA evaporator takes the first cycle aqueous raffinates from reprocessing, which bear over 99% of the fission products and minor actinides, and concentrates them by a factor of 20-100 for interim storage in high integrity, double-walled stainless steel tanks. The stored concentrate is subsequently converted by vitrification into monolithic glass blocks encased in stainless steel cylinders, which are suitable for long term storage and ultimate disposal.

The salt evaporator deals principally with solvent wash raffinates. The organic solvent (TBP/OK) used in the main chemical separation process becomes contaminated and degraded over time, so is chemically washed and recycled into the process in order to minimise waste arisings. The solvent wash raffinates carry a heavy burden of sodium hydroxide and carbonate from the washing process, so are unsuitable for evaporation with other salt-free MA effluents. Hence they are neutralised and concentrated in a separate salt evaporator, the concentrate being delay-stored for ruthenium decay prior to treatment in EARP.

There are some aqueous effluents which are too voluminous for evaporation but contain concentrations of certain radionuclides, especially actinides, which make them unsuitable for direct sea discharge. Most of these streams arise in the Magnox plant and are acidic iron-bearing streams. This feature has been used in the development of the Enhanced Actinide Removal Plant (EARP - see Fig 6). The streams are neutralised with caustic soda to produce a ferric hydroxide floc which carries with it virtually all the actinides. Addition of small quantities of specific chemicals also improves beta decontamination. The floc as first produced is too voluminous and needs to be de-watered for economic storage and disposal. This is achieved by ultrafiltration, giving typical concentration factors of ~500. The concentrated floc is encapsulated in cement ready for disposal and the permeate from ultrafiltration, being very low in radioactivity, is discharged to sea. Only a small number of

streams from THORP are fed to EARP; these include certain solvent washes from the plutonium purification cycle, Multi-Element Bottle (MEB) flushings from the Feed Pond and salt evaporator concentrate (which contains arisings from Magnox and THORP).

Waste solvent arisings are minimised by the use of solvent washing and recycle. However the two sources of arising in THORP are the HA cycle solvent purge and float-off solvents arising from end-of-campaign washouts. Waste solvents from Magnox and THORP reprocessing are currently stored pending completion and commissioning of the solvent treatment plant (STP - See Fig 7). The process at the heart of STP is alkaline hydrolysis. Boiling the spent solvent with concentrated caustic soda breaks it down into three phases: an organic kerosene phase which can be burned; a sodium dibutyl phosphate phase (Na DBP) which is low enough in contaminants for sea discharge; and an aqueous sodium hydroxide phase containing the vast majority of the radioactivity which can be combined with aqueous wash streams from the pretreatment stages and fed to EARP for clean up and discharge.

## PREDICTED DISCHARGES

### Gaseous

A comparison of the flowsheeted discharges with the latest Discharge Authorisation is given in Table 1, based on the design throughput of 1200t(U)yr of reference fuel. For all radionuclides with a defined limit, there is a reasonable margin between the flowsheeted discharge and the authorised limit. In terms of the Critical Group impact, the predicted dose under flowsheet conditions is 22μSv/yr, against the target dose of 50μSv/yr.

### Liquid

As mentioned previously, THORP does not have its own Liquid Discharge Authorisation so Table 4 compares the predicted THORP discharges, both direct and via downstream treatment plants, with the total site Discharge Authorisation. In general, it can be seen that the predicted discharges are only a small percentage (generally <10%) of the total site allocation. For the Critical Group impact, the predicted dose is 36μSv for local seafood and shellfish eaters. This group is distinct from the aerial discharge Critical Group, so can be compared in isolation with the target of 50μSv.

**Table 4 - Comparison of THORP Liquid discharges with Total Site Authorisation (TBq/yr)**

| Radionuclide | Total Predicted THORP Discharges | Site Authorisation |
|---|---|---|
| H-3 | 6,972 | 31,000 |
| C-14 | 0.497 | 20.8 |
| Co-60 | 2.37 | 13 |
| Sr-90 | 0.844 | 48 |
| Ru-106 | 18.2 | 63 |
| I-129 | 1.40 | 2 |
| Cs-137 | 6.07 | 75 |
| Pu (alpha) | 0.045 | 0.7 |
| Total alpha | 0.096 | 1 |
| Beta-5 | 30.6 | 400 |
| Critical Group dose = 36 μSv/yr | | Target dose = 50 μSv/yr |

## CONCLUSIONS

1 BNFL's Thermal Oxide Reprocessing Plant incorporates a variety of modern techniques for the treatment of gaseous and liquid effluents.

2 For off-gases, the design target has been met by subdivision of the arisings into discrete treatment trains, viz: dissolver off-gas, central off-gas, gloveboxes, cells and caves, and building ventilation.

3 Each stream is treated by a combination of process steps appropriate to its composition and the required degree of decontamination.

4 Development work on key parts of the off-gas treatment system, supported by theoretical modelling, has ensured that plant design has a firm foundation.

5 All active streams are discharged, after treatment, from a 125m stack whose effective height has been demonstrated to be 92m.

6 Modelling work predicts that the critical group dose from THORP aerial discharges will be 22μSv compared with a design target of 50μSv.

7 The treatment of liquid effluents has been simplified by the use of salt-free reagents and recycling of waste streams within the process.

8 Liquid effluents are either treated by dedicated equipment in THORP or combined with other streams for treatment in common site facilities, prior to sea discharge.

9 The treatment process used depends on the volume, nature and specific radionuclide content of the effluent.

10 The net result is a predicted dose to the marine critical group of 36μSv, against the design target of 50μSv.

11 The individual radionuclide discharges from THORP generally will make only a small contribution to the total site liquid discharges.

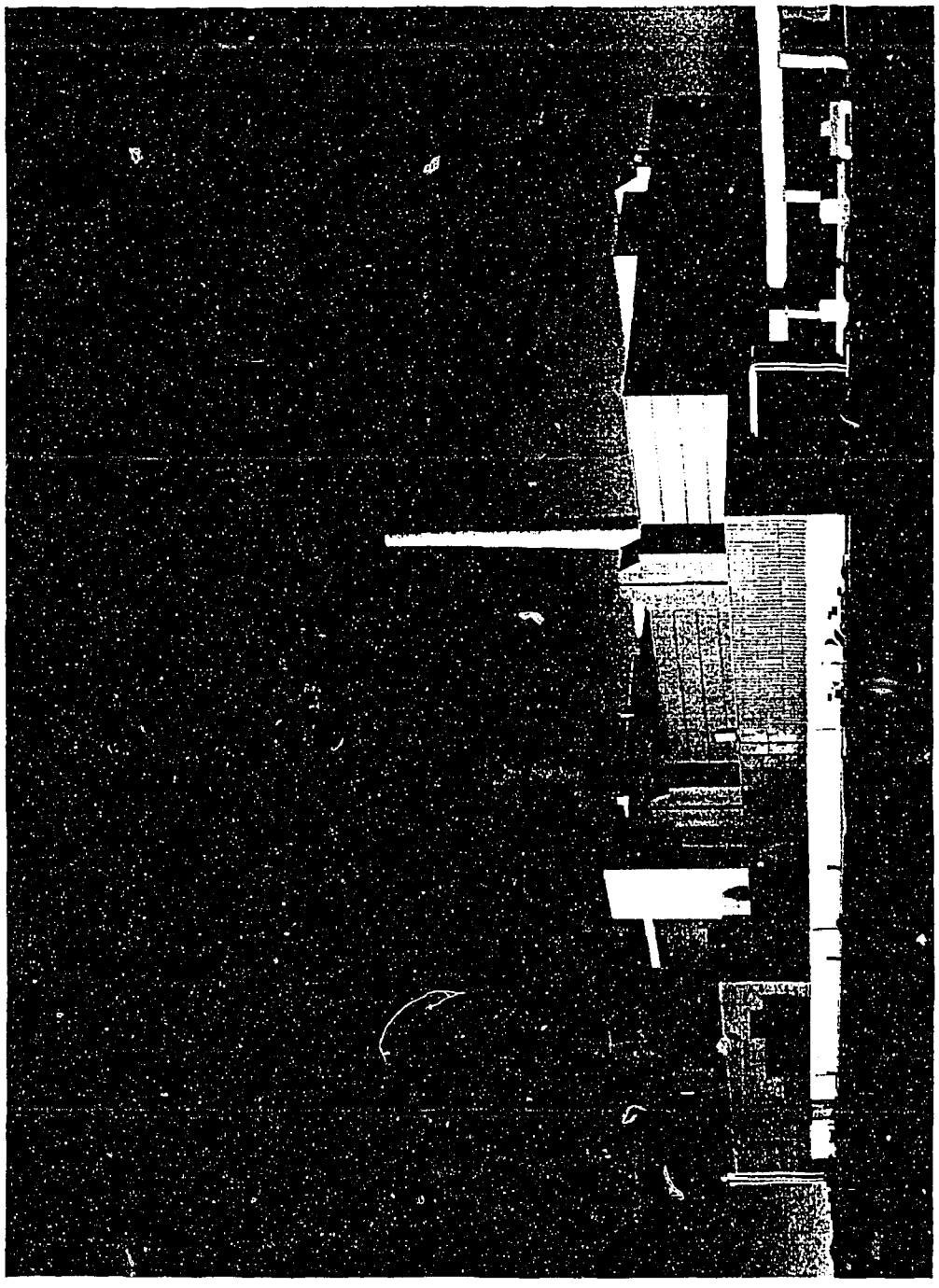

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Figure 1 THORP building

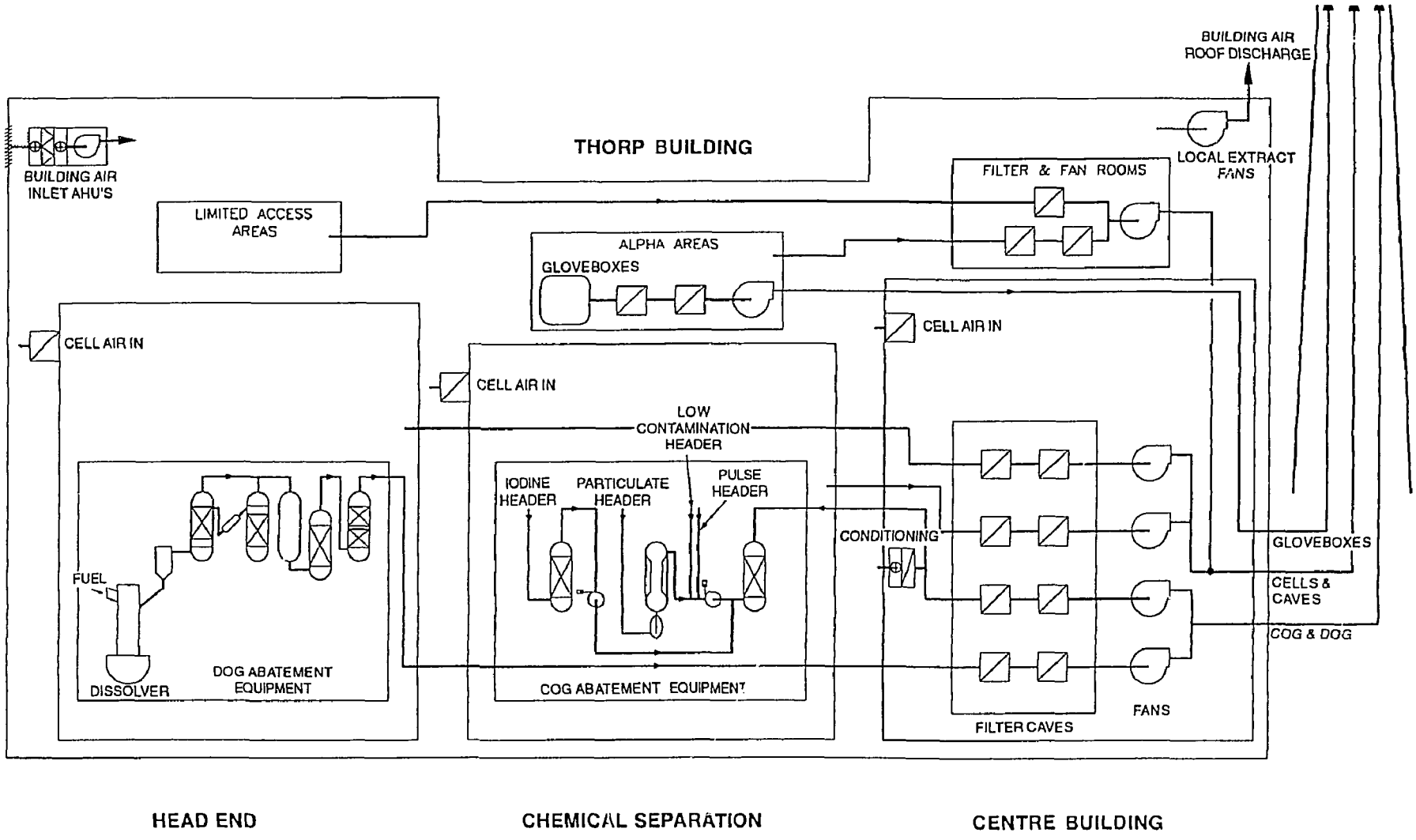


**Figure 2** Schematic of THORP active ventilation systems

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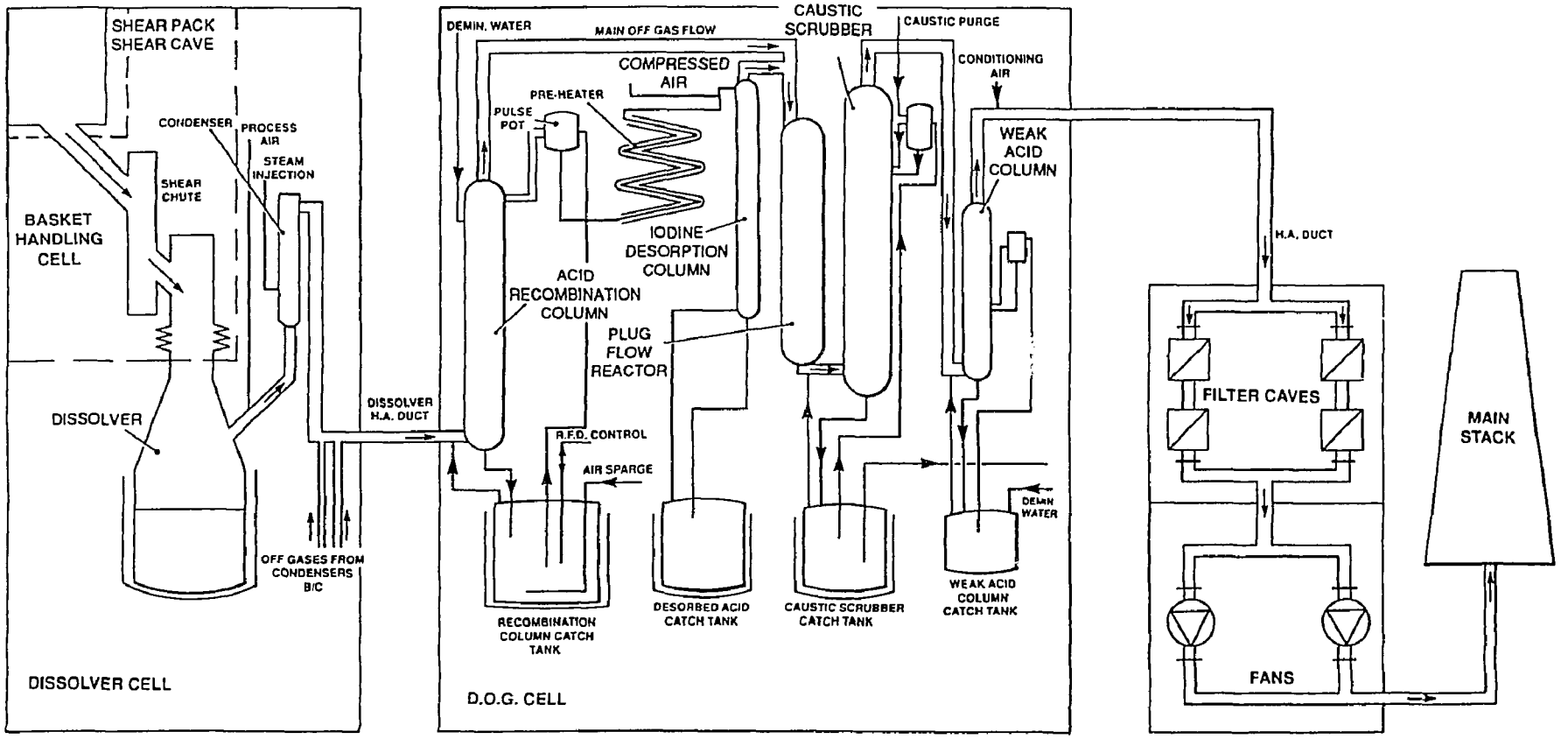


**Figure 3 Dissolver off gas (DOG) extract system**

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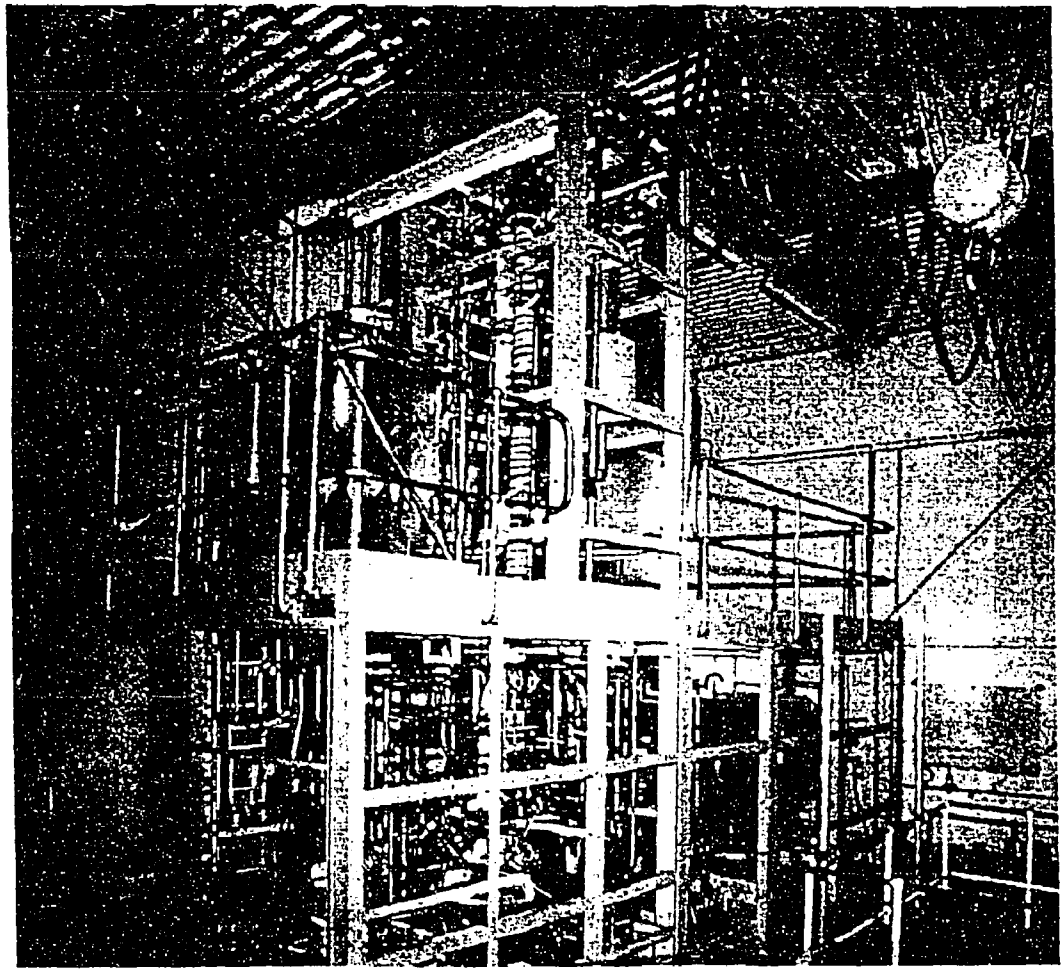

Figure 4 Dissolver off gas pilot plant

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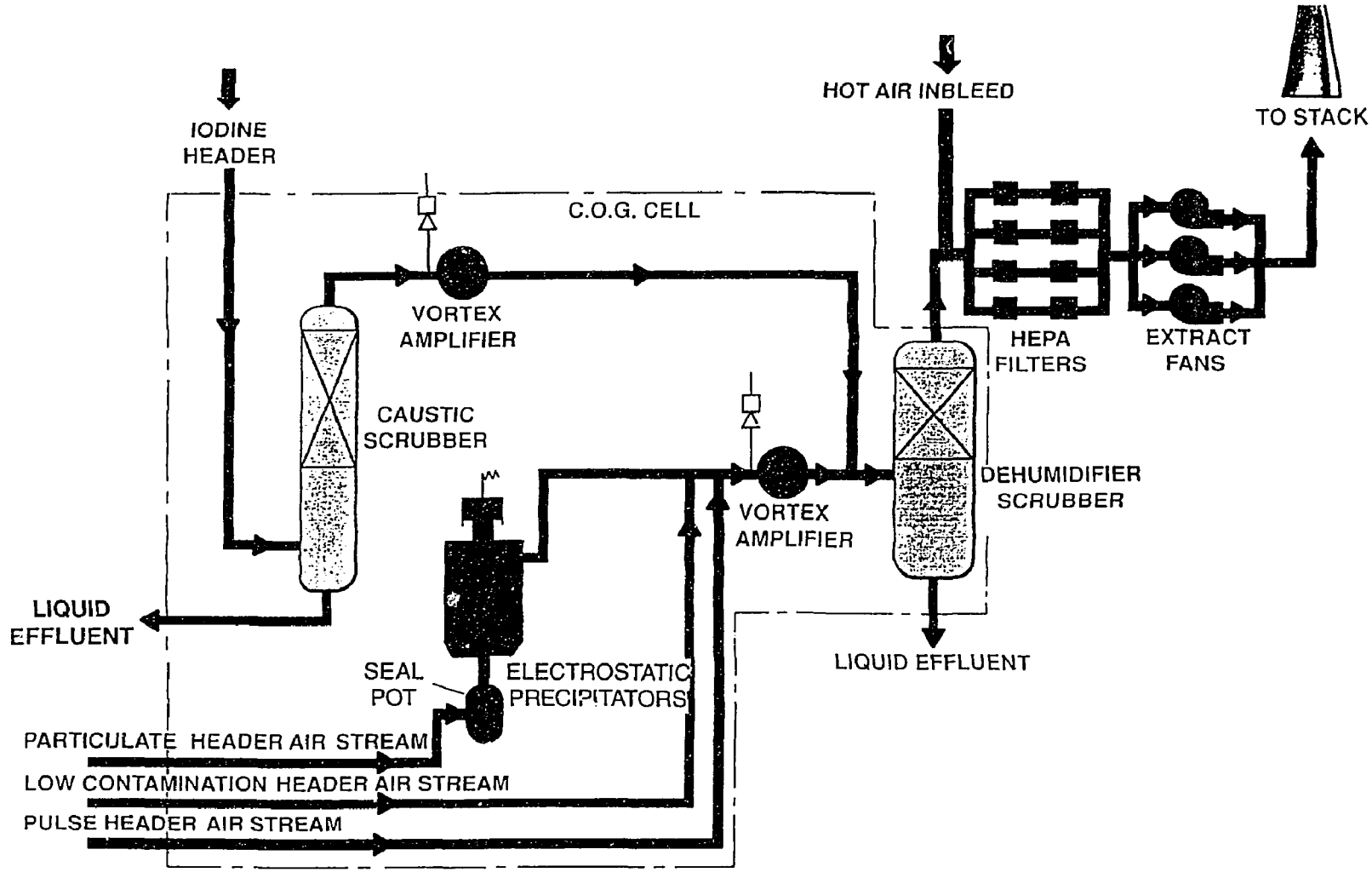


**Figure 5** Central off gas (COG) extract system

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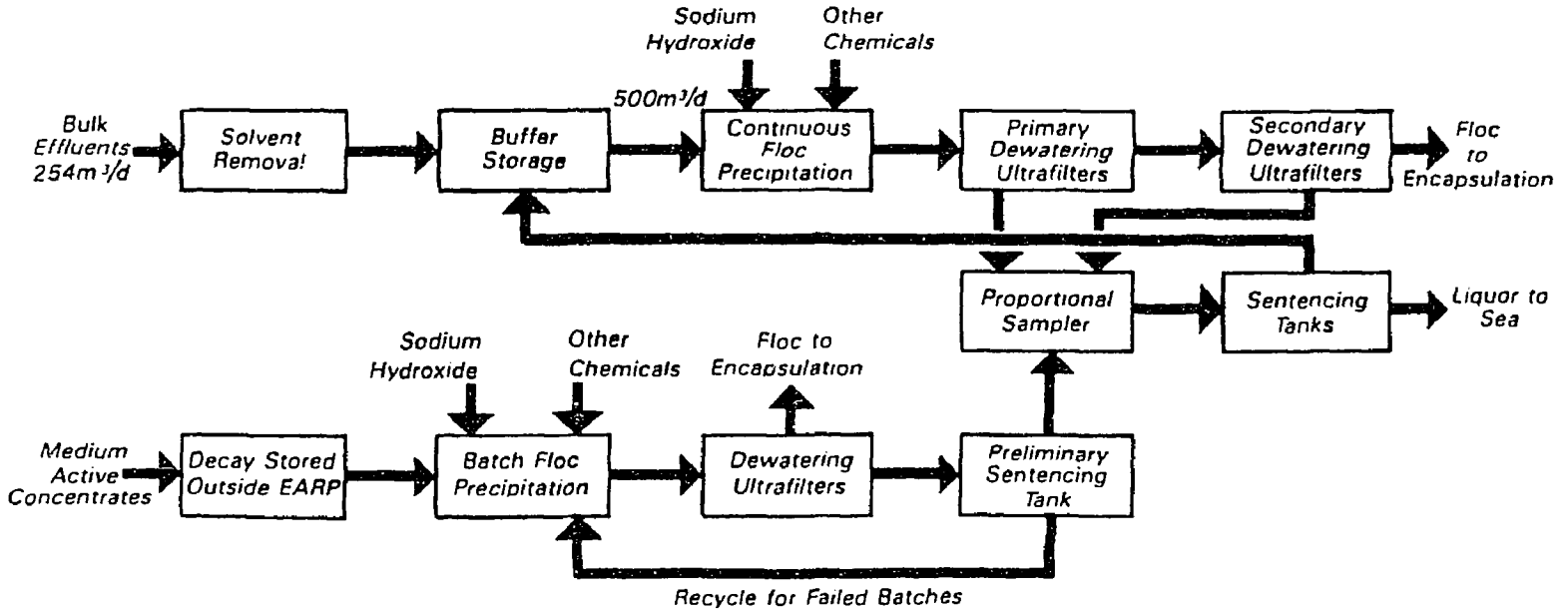


**Figure 6 EARP - Simplified Process Flow Diagram**

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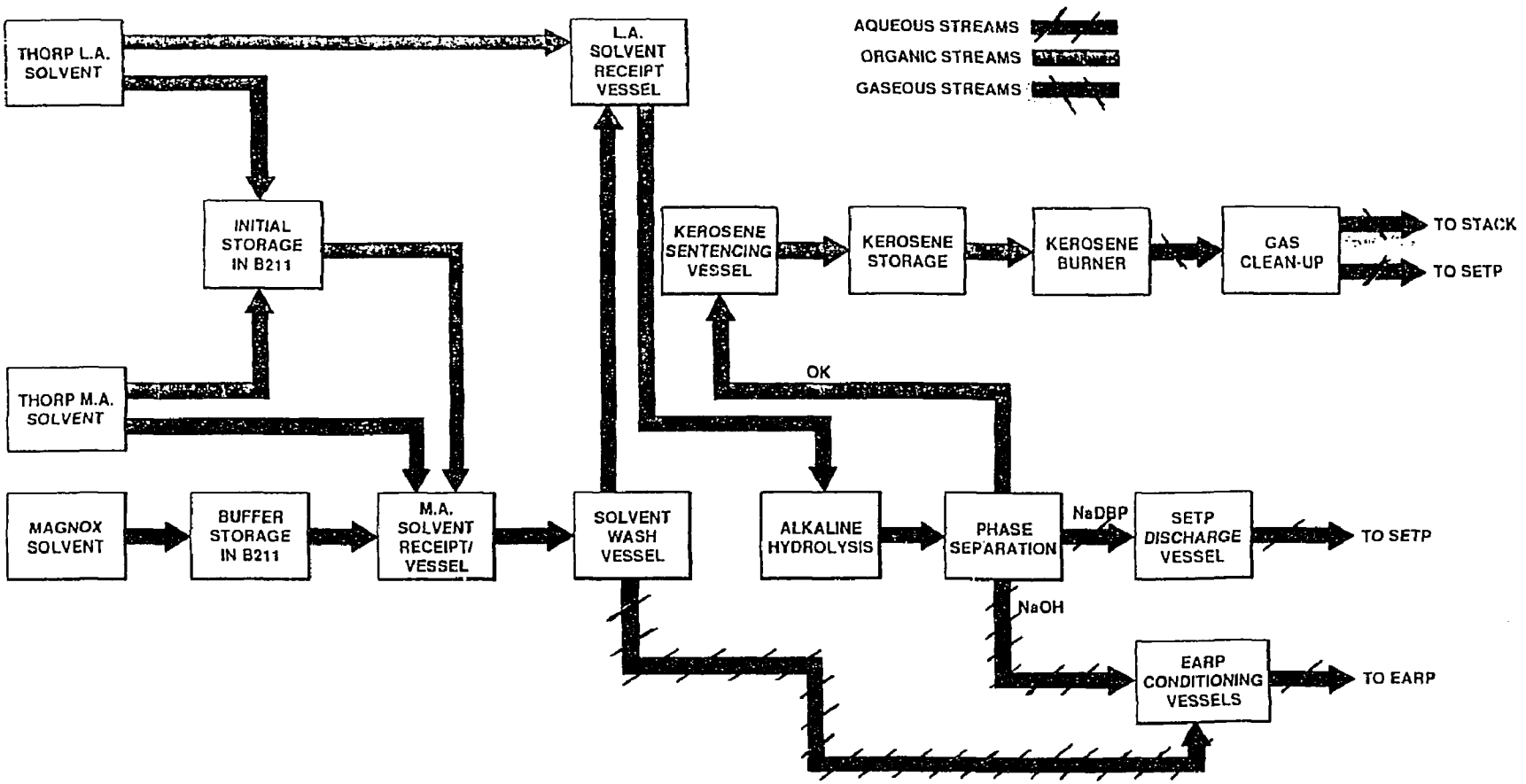


Figure 7 STP Block Flow Diagram