BNFL'S OBJECTIVES AND ACHIEVEMENTS IN THE REDUCTION OF RADIOACTIVE DISCHARGES FROM REPROCESSING PLANTS

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ABSTRACT

The reprocessing of irradiated nuclear fuel has been carried out at Sellafield in Cumbria since 1952. Since 1971 British Nuclear Fuels plc (BNFL) has been responsible for operating the site and providing fuel cycle services to both British and overseas customers. BNFL is also responsible for the treatment of any wastes generated as a result of providing these services. Some liquid effluents are discharged to sea after treatment. In order to minimise the amount of radioactivity discharged to the environment the Company is introducing new plant which, by the early 1990's, will reduce annual Sellafield discharges to about 0.74 TBq (20 Ci) alpha and 300 TBq (8,000 Ci) beta. This paper describes the regulatory framework within which BNFL operates, the reduction of Sellafield discharges and the three principal processes by which this reduction is being achieved.

INTRODUCTION

Reprocessing operations commenced at Sellafield in 1952 when the first reprocessing plant was commissioned. A second facility, brought on line in 1964 to process irradiated uranium metal fuel from the UK civil Magnox reactor program, is expected to continue in operation until the early years of the next century. A further reprocessing plant, to treat irradiated oxide fuel, the Thermal Oxide Reprocessing Plant (THORP), is being constructed for operation in 1992.

Prior to reprocessing, irradiated fuel is stored in water-filled ponds, and these storage and reprocessing operations result in the generation of a number of radioactive liquid effluents. The highly active liquid effluents are stored in cooled, high integrity, stainless steel tanks and will, with the commissioning of the Windscale Vitrification Plant, be converted into glass cylinders for long term storage and disposal. Medium active liquors are evaporated to reduce their volumes and are decay-stored to permit the decay of short-lived isotopes. Although these concentrates used to be discharged to sea after a period of storage, since the early 1980s no such discharges have been made. Low level active effluents are discharged to sea, well within the authorised limits for the Sellafield site.

For many years, BNFL has adopted a policy of reducing the levels of activity discharged to sea and significant improvements have already been made. In 1985, the Site Ion Exchange Plant (SIXEP) was brought into operation to treat water from the fuel storage ponds, mainly in order to remove caesium and strontium. The evaporation and decay-storage of liquid wastes from solvent extraction has played a major part in reducing site discharge and, also in 1985, with the commissioning of the Salt Evaporator, this was extended to include the evaporation of sodium salt-bearing liquors from solvent washing operations.

The storage of medium active and salt concentrates, which currently amount to about 700m^3 per year, can, however, only be regarded as an interim measure and further treatment is required. Additionally, one fifth of the low active effluent not treated by SIXEP, amounting to about 250m^3 per day, accounts for the majority of the activity currently discharged to sea. Treatment of this effluent would substantially reduce the total site discharge of activity to sea and a plant to remove alpha activity (actinides), and some beta activity, from this stream and from stored medium active concentrates is being designed. This plant, called the Enhanced Actinide Removal Plant (EARP), is scheduled to be operational by 1992.

DISCHARGES FROM THE SELLAFIELD SITE

Radioactive discharges from the Sellafield Site have always been subject to authorisation by the appropriate UK regulatory bodies which, since 1960,

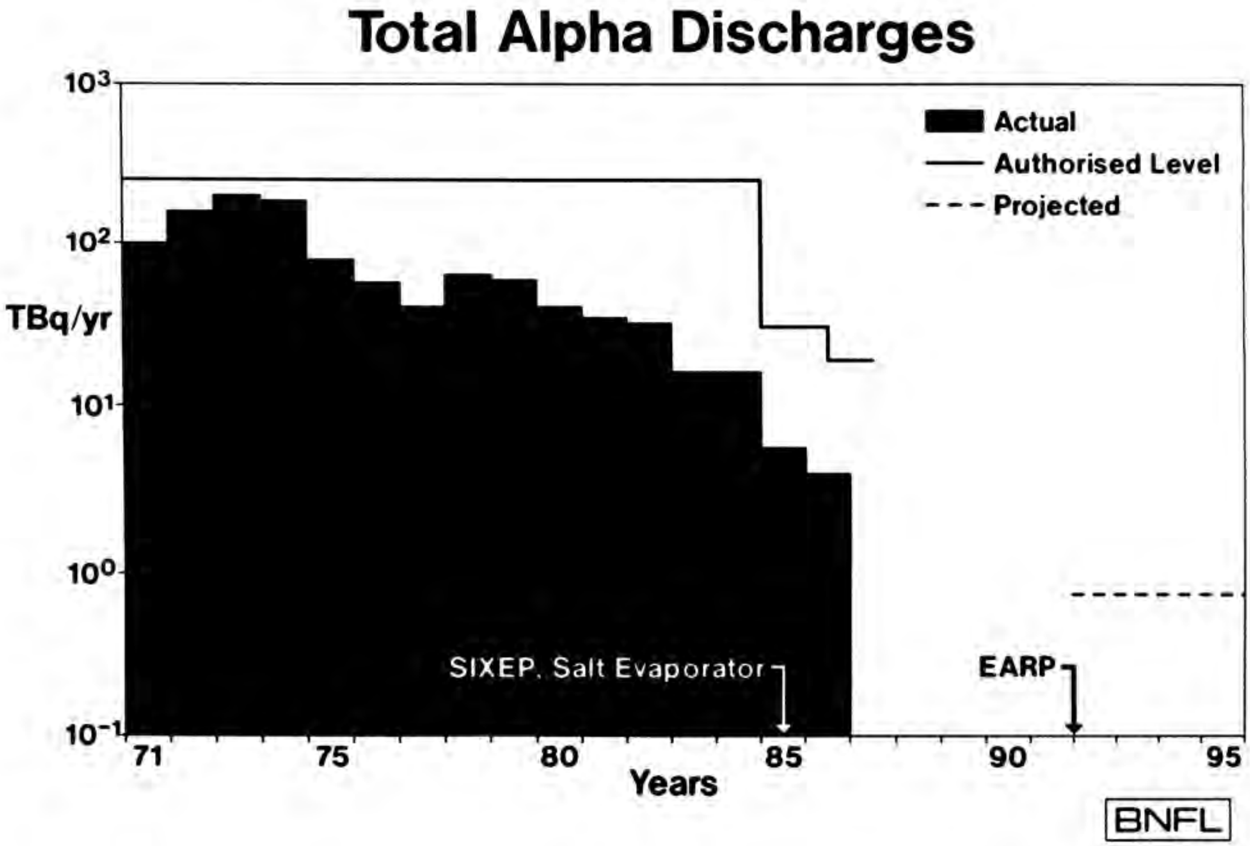

Fig. 1. Total Alpha Discharges.

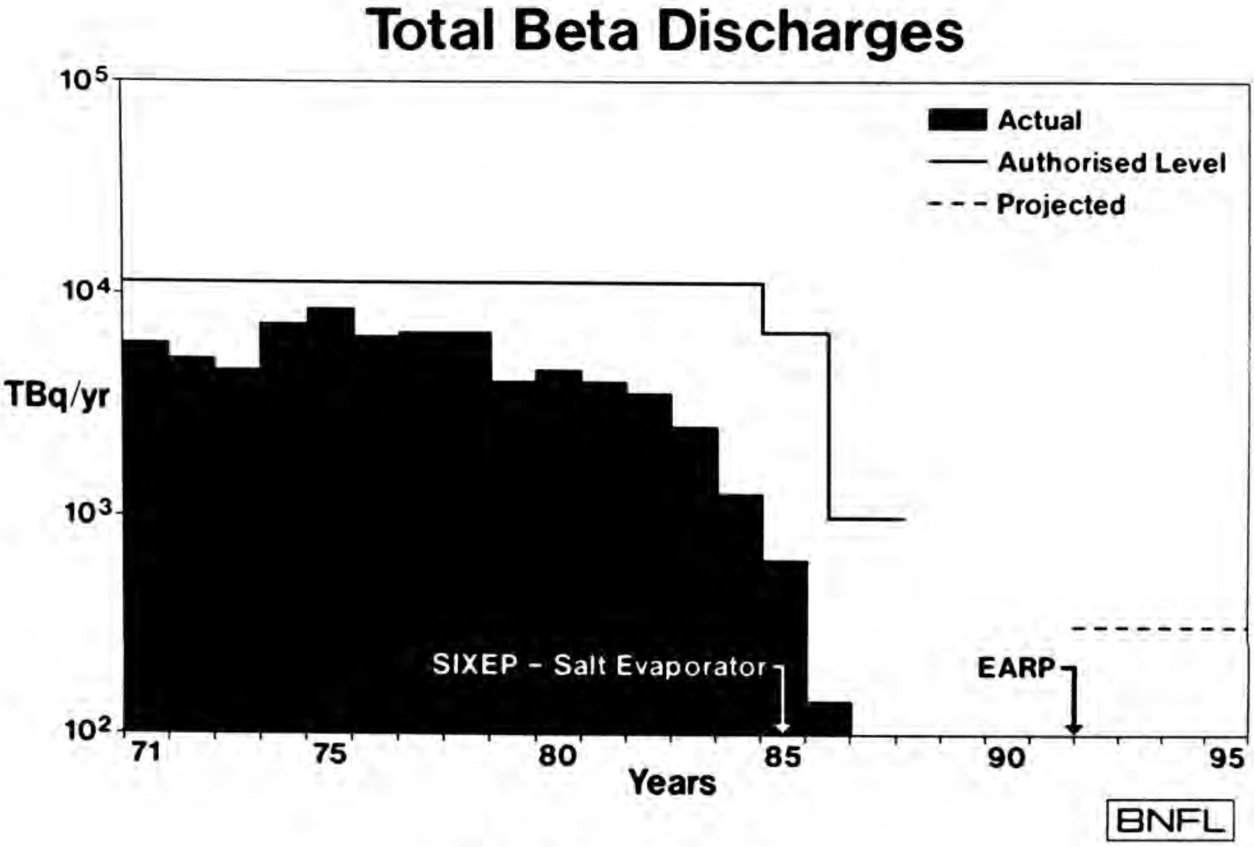

Fig. 2. Total Beta Discharges.

have been the Department of the Environment (DoE) and the Ministry of Agriculture, Fisheries and Food (MAFF). Within this framework BNFL has formulated its own policy objectives and targets for control of discharges and radiation protection of the public. These are as follows:

1. That discharges to the environment should be kept within authorised limits.

2. That discharges should be controlled so that the resulting radiation doses to the most highly exposed members of the public (the "critical groups") are well within the limits recommended by the International Commission on Radiological Protection (ICRP) and that the principles of radiological protection, including ALARA (As Low As Reasonably Achievable), promulgated by ICRP, are taken fully into account.

3. That, over and above ALARA considerations, the radiation doses to the most exposed members of the public should be restricted to a target of less than about 0.5 mSv per year (ie 10% of the limit recommended by ICRP for any single year's exposure).

The regulatory bodies keep the authorisation under review in the light of advice from ICRP, the UK's Radioactive Waste Management Advisory Committee (RWMAC) and the National Radiological Protection Board (NRPB), and of discharge reductions achieved. As a result, an ALARA clause was introduced in 1983 and lower annual limits and restrictions on solvent and solids content of discharges were introduced in 1985. An extensive, and more restrictive, revision came into effect in July 1986, following the introduction in 1985 of the new effluent treatment plants mentioned previously. These plants were designed to enable achievement of the above target but changes in critical group habits and parameters in the meantime have delayed this. The new authorisation also requires the Company to use the best practicable means (BPM) to limit the amount of activity discharged.

Figures 1 and 2 show the past and projected Sellafield discharges from BNFL's formation in 1971 up to 1995, by which time all the effluent treatment plants currently planned will be in operation. The "total beta" figures given in Figure 1 refer to a specific method of estimation which effectively excludes low energy beta emitters, most significantly tritium and Pu241. However, both these radionuclides are measured in effluents and taken fully into account in the assessment of radiological impact. Discharges have at all times remained within annual authorised limits.

The impact of radioactive discharges into the environment, expressed in terms of the resulting radiation dose to members of the public, depends on many factors, amongst which the quantities discharged, environmental dispersion, food chain concentration, public habits and consumption patterns, and human metabolism are the most important.

In assessing the significance of future discharges, extensive use is made of past discharge and environmental data, together with limited mathematical modelling, to establish relationships between discharge rates and levels of radioactivity in the environment. Such relationships, established independently by the Company and regulatory bodies, are used to guide the design of plant towards the achievement of the Company's objectives.

Prior to the mid 1970s the critical group was based on the intake of Ru 106 as a result of the consumption of laverbread. However, the peak in total beta discharges in 1975 was due to discharges, primarily Cs 137, from the fuel storage ponds as a result of leaching through corroded Magnox fuel cladding. The critical group thus became people eating large quantities of local fish and crustaceans (about 80 kg and 15 kg per year, respectively). Radiation doses rose to nearly 2 mSv per year. To conform with BNFL's own criterion of limiting critical group doses, reductions in activity discharges were effected by introducing local ion exchange units (for caesium) and by the re-routing of an alpha-bearing stream to highly active storage.

The combined effect of these and other measures was to progressively reduce total beta discharges to some 1190 TBq in 1984, and with the recent commissioning of SIXEP and the Salt Evaporator, described in detail later, there was a further reduction of about 600 TBq in 1985 and about 130 TBq in 1986, the latter reflecting the good performance of the Salt Evaporator and SIXEP plants in their first full year of operation.

By 1981, alpha discharges had been reduced to about 30 TBq per year. The radiation dose to the public resulting from alpha discharges, due mainly to high rate consumption of molluscs, was assessed to be less than 0.2 mSv per year. However, in subsequent years, the regulatory bodies reported increasing consumption of molluscs by the critical group, rising from an initial value of about 2 kg per year, through 6.5 kg per year, to a highest value of about 16.5 kg per year, reported in late 1983. In addition, the NRPB advised in early 1983 that the assessed radiation dose resulting from ingestion of plutonium in environmental material should be increased by a factor of 5. These retrospective changes created a new critical group whose dose originates almost entirely from uptake of actinides due to high rate consumption of molluscs. Their assessed annual radiation dose reached a figure of about 3.5 mSv in 1981 (about 70% of the ICRP dose limit).

During this period of change, in October 1983, the Company announced measures to reduce alpha discharges further to about 7.4 TBq/yr. These measures are now effective, and operating as expected, with the result that the radiation dose to the critical group in 1985 was below 2 mSv per year and falling. Alpha discharges were about 6 TBq in 1985 and about 4 TBq in 1986.

In 1984, further measures were announced, endorsed by RWMAC, and approved by the Government who considered that they complied with the 1984 recommendations of the Paris Commission. These new measures, which include EARP, are expected to be complete by 1992. They are expected to result in discharges of about 0.74 TBq alpha and 300 TBq beta per year. In the meantime, discharges will remain at about current levels, with actual levels being determined by reprocessing throughput and the performance of effluent treatment plants. Radiation doses to the critical group are expected to fall to about 0.1 mSv per year by around the turn of the century. The exposure of the general public will be much less than this.

MAJOR TREATMENT PLANT AND PROCESSES

Site Ion Exchange Plant (SIXEP)

Spent fuel delivered to Sellafield must be stored before it is reprocessed to separate reusable materials and waste. The spent fuel is highly radioactive and still produces decay heat. It is, therefore, necessary for the storage facilities to provide radiation shielding and a system for heat removal. It is, of course, also essential that the fuel be readily handled in the store. The most flexible form of storage has been found to be in water-filled ponds, generally of the order of thirty to forty feet deep. The water provides both the necessary radiation shielding and the heat removal medium.

However, while in storage, some corrosion of the fuel takes place and this leads to the production of radioactive sludges and the presence of radioactive ions in the storage water. In the case of Magnox fuel (clad with magnesium alloy) which has to date accounted for most of the fuel from the British Nuclear Power Program, the amount of corrosion is limited as far as possible by maintaining the storage water at pH 11.5 by dosing with sodium hydroxide, and by a low bulk water temperature. In addition, the concentration of non-radioactive ions, such as chloride, sulfate and silicate, all of which to a greater or lesser extent effect the corrosion of Magnox, is maintained at very low levels. Such corrosion as does take place results in the release of caesium 137, caesium 134, and strontium 90 to the pond water. This, if untreated, would result in radiation exposure to the plant operators.

In order to enable the concentrations of both radioactive and non-radioactive ions to be limited to acceptable levels, water is continually purged from the ponds. The water volume which must be purged to maintain the correct limits has been found to be approximately six percent of the pond volume per day and this amounts to over 3000m^3 of purge for all the current Sellafield ponds. The volume purged is made up with clean demineralised water dosed to the correct pH value.

The SIXEP plant was constructed, at a cost of about \$180M, to remove the caesium and strontium from these pond purges. The plant is also required to remove any sludges carried over with the various purges. A study of the methods of removing caesium and strontium available for large scale application showed that ion exchange in pressurized columns was the most appropriate method. Consideration of the size range of sludges to be removed showed that these too would be removed on ion exchange columns. However, this would have led to a pressure build up and the need to backwash the columns at frequent intervals. Backwashing ion exchange columns disturbs the bed of material and adversely affects the performance so sand pressure filters were installed for sludge removal. In terms of plant flexibility, it was concluded that there was a justifiable case for making the ion exchange columns and sand pressure filters identical in design and to provide pipework to allow the columns to be operated in any desired mode. This led to a process flow diagram as shown, in a simplified form, in Fig. 3.

Feeds from the various ponds and processes are collected in a reception tank before being fed to the first stage of the treatment process: the removal of sludges and suspended solids present in the liquor. This is necessary for two reasons: firstly, as mentioned before, solids not removed before the ion exchangers are liable to be trapped on these columns, causing high pressure drops which could only be reduced by backwashing, thus affecting the actual ion exchange process, and secondly any magnesium solids not removed before pH reduction would go into solution and produce ions which would compete with the caesium and strontium.

In order to maximize the efficiency of the sand pressure filters, in terms of both the actual percentage solids removed and the quantity of liquor treated between backwashes, it was established that a low molecular weight polyelectrolyte, such as Nalfloc N7607 or Magnafloc 1597, should be used, that the feed temperature should ideally not fall below 20°C and that 10 mg/l of silica in the feed was desirable. At the end of each duty cycle, the sand pressure filters are backwashed and the collected sludges are transferred to 1000m^3 storage tanks.

Liquor from the sand pressure filters passes to the carbonation tower. While evaluating candidate ion exchange materials, it was noted that some promising materials, although stable at pH values of up to 10.0, were subject to silica leach from the crystalline matrix at higher pH values, thus weakening the materials. It was therefore decided to operate the ion exchange columns at pH 8.0 and to reduce the feed pH value from 11.5 using a carbon dioxide counter flow carbonating tower. This method was selected as it has been shown that the lowest pH value which can be achieved is higher than the value at which acid attack of, or activity leaching from, the ion exchanger can be achieved.

Liquor is then fed to the ion exchange columns. When radioactive liquors are treated by ion exchange, it is not usually possible to regenerate the ion exchanger at the end of its operating cycle, as in a conventional plant, since this leads to an effluent containing concentrated activity which cannot be disposed of to drain in the normal way. Consequently, the ion exchanger is hydraulically discharged from the columns at the end of each operating cycle, for tank storage, and replaced with a new charge.

Clearly, storing spent ion exchanger is a costly process and there is a very real incentive to find materials which are highly selective for caesium and strontium, thus increasing the volume of liquor treated before the end of the cycle is reached. Additionally, the storage of spent ion exchanger can only be regarded as an interim measure and some form of permanent immobilization is required. The most straightforward and cost effective method of immobilization is by encapsulation in a cement-based matrix.

Extensive testing identified a material which would satisfy both the above criteria and thus the natural zeolite, clinoptilolite, was specified for SIXEP. Typically, at 30,000 bed volumes of liquor treated, decontamination factors (DF) of the order of 15 are being achieved. In practice, two ion exchange columns will be used and, during the first 20 months of SIXEP operations, a DF of about 900 has been achieved for caesium. Again, in order to minimise the volume of spent ion exchanger arisings, work has been carried out to optimise a system of column reversal such that each charge of material is used first in the lag column followed by use in the lead column. In this way it is anticipated that about 40m^3 of ion exchanger will be used per year.

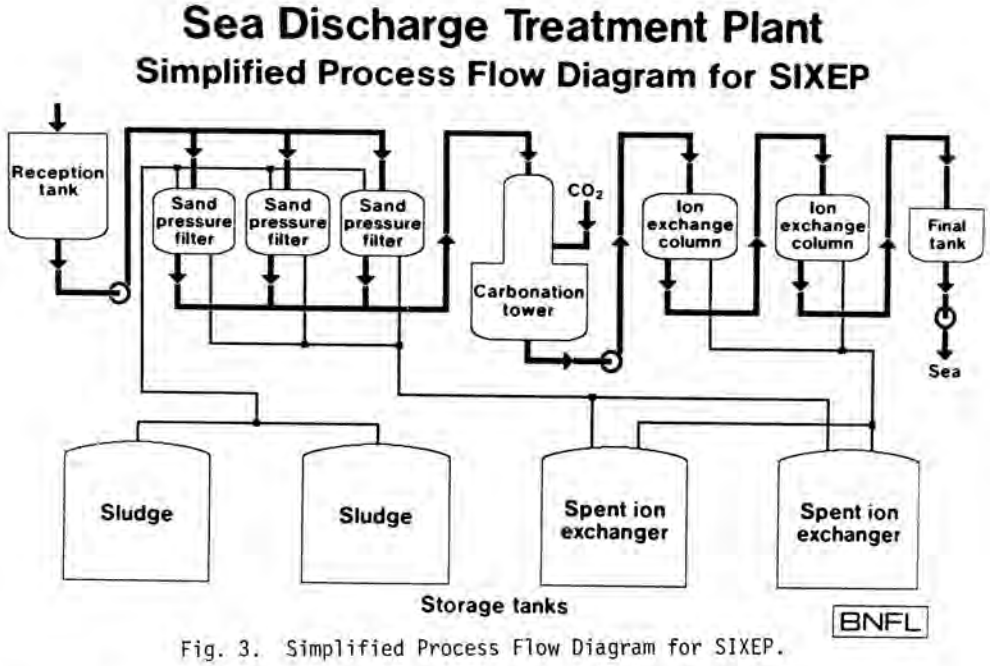

Fig. 3. Simplified Process Flow Diagram for SIXEP.

Work has shown that small quantities of spent ion exchanger left behind when the bed is hydraulically discharged has a very marked effect on the DF obtained from the subsequent charge in that vessel. This led to the need to design and develop very carefully the actual ion exchange columns so that a minimum residue is left.

Having passed through the ion exchange columns the liquor is passed through a proportional sampler to a final break tank from where it is filtered, sampled and pumped into the sea.

The plant has been designed to be extremely flexible. The universal pressure vessels used for both the sand pressure filters and the ion exchange columns have been produced to a common design and are, in fact, dual purpose. The only major design change required to achieve this commonality was to increase the original height of the sand pressure filter vessels. From Fig. 3, it can be seen that connections have been provided such that any one of the sand pressure filters can be used as an ion exchange column if required. The carbonation tower maintains the pH of liquor entering the ion exchanger columns at a level low enough to prevent "weakening" of the clinoptilolite. Connections have been installed to enable the carbonation tower to be by-passed, should improved ion exchange materials which require a higher pH be identified.

Figure 4 shows a plan of the SIXEP building. The four $1000m^3$ storage tanks can be seen at the top left and the sand pressure filters and ion exchange columns at the bottom left. A central spine contains the main valves and pumps associated with the process.

The Salt Evaporator

From the mid-1970s, it was recognised that the direct discharge of sodium salt-bearing liquors, from solvent washing operations, was a major contributor to the level of Ru 106 in the environment. Since Ru 106 has a short half life (about 1 year) it was decided to decay-store these liquors, after concentration by evaporation, to permit most of the Ru 106 to decay before discharge to sea. Because of the corrosive nature of these salt-bearing liquors a new dedicated evaporator was required.

During the design and construction stage, changes in the way in which environmental impact was perceived resulted in modifications to the plant. The first was the installation of a caustic scrubber to remove radioactive iodine which would, otherwise, be vented to atmosphere. The second, resulting from the changes in the critical group, noted earlier, and the consequent importance of reducing the level of actinides discharged, was to route plutonium-bearing streams, which had previously been discharged to sea, to evaporation and storage. In particular, raffinates from solvent washing during the plutonium purification cycle where routed to the Salt Evaporator. A further modification was the installation of "weirs" in the feed liquor storage tanks to permit the periodic removal of any free-phase solvent carried over into the tanks.

Figure 5 shows a simplified process flow diagram. The feed liquors destined for the Salt Evaporator enter the storage tank via the gravity flow active pipebridge. A distributor directs the incoming liquor into either of the two storage tanks which are cooled to remove the latent heat of steam ejector transfer operations. Each tank has an ejector for transferring liquor to the other. The tanks are also interconnected via an overflow line.

The alkaline feed liquors are ejected from either of the storage tanks, via a breakpot and the constant volume feeder (CVF), to the conditioning vessel. This is a stirred steam-heated vessel in which the feed is acidified to pH2 and heated to 90°C. The conditioned feed passes to the steam strip column where residual solvent is removed. Solvent vapours and steam from this column are condensed and passed to a monitoring system and sent to the site effluent system. The steam stripped liquor passes via a cooler into the vacuum evaporator system.

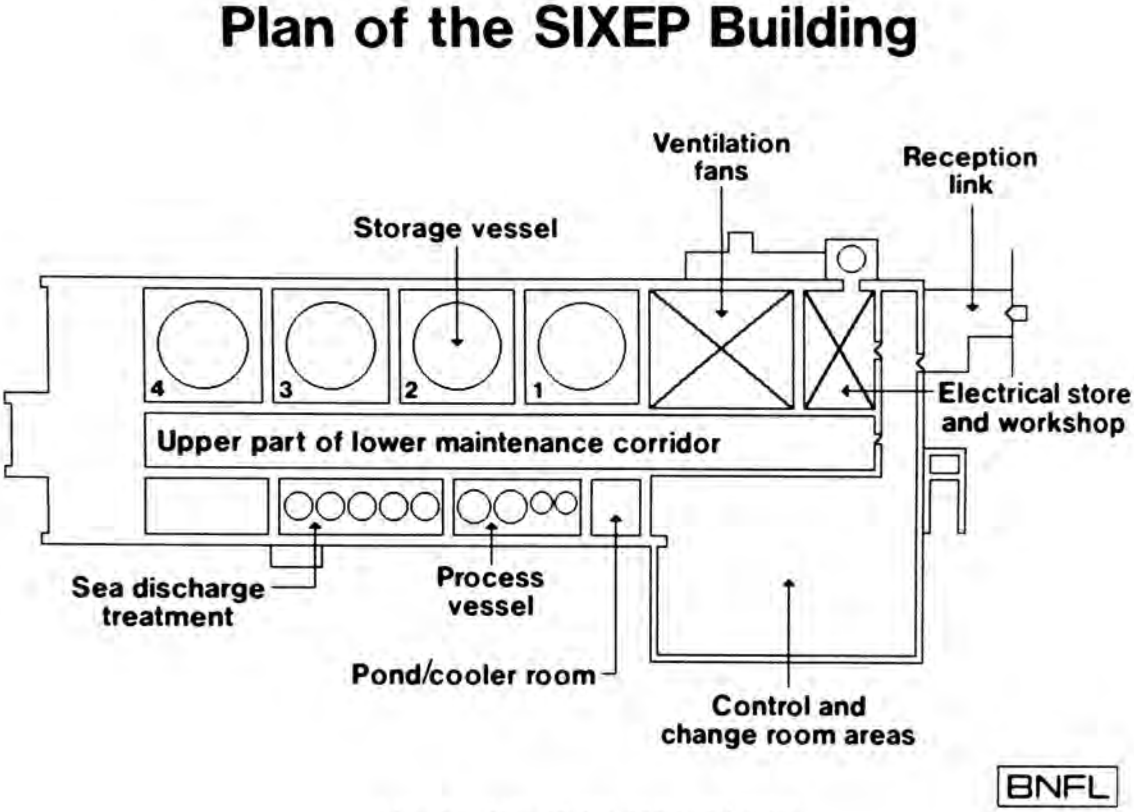

Fig. 4. Plan of the SIXEP Building.

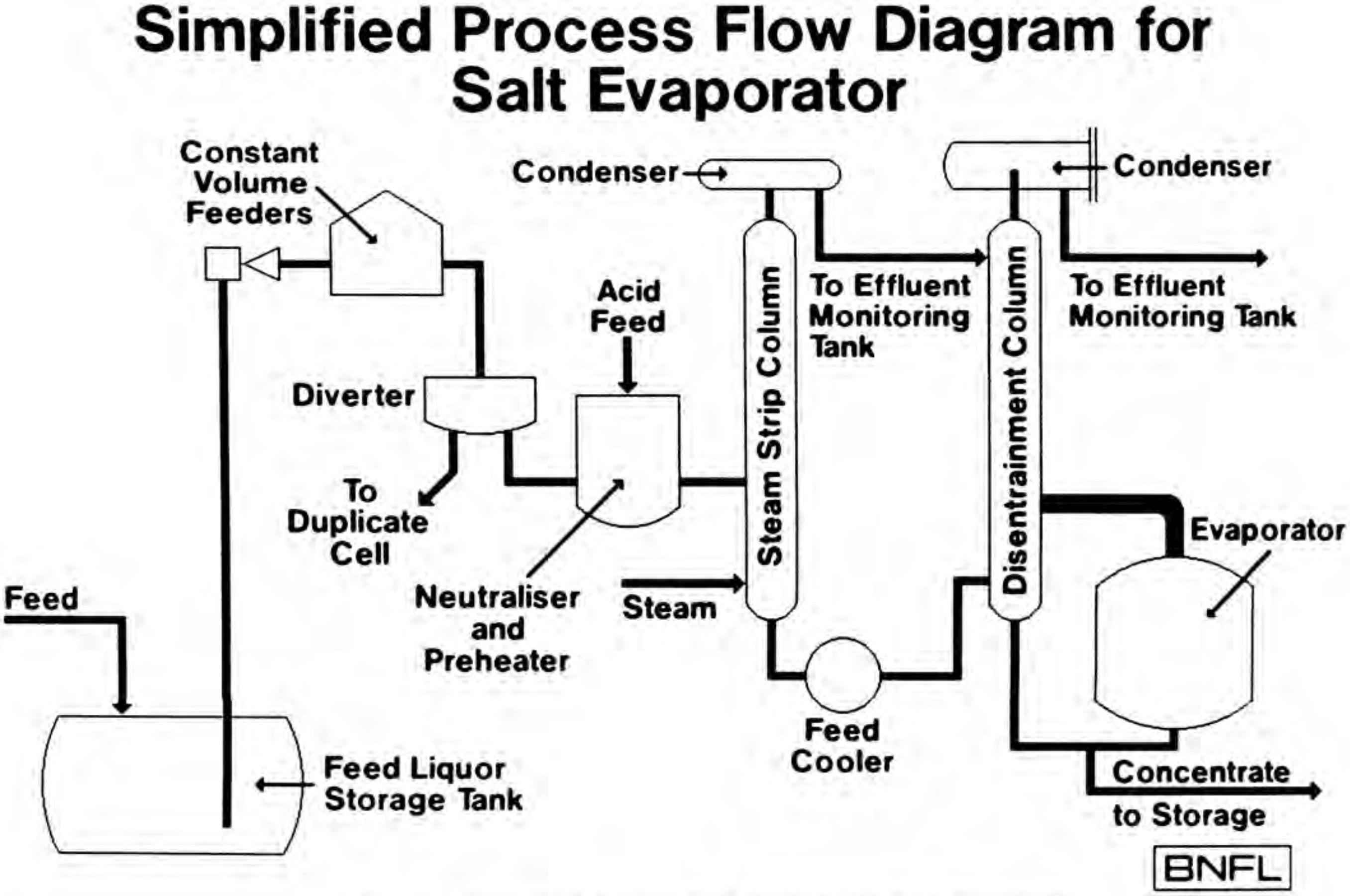

Fig. 5. Simplified Process Flow Diagram for Salt Evaporator.

The feed enters the disentrainment column and flows, via the recirculating leg, to the evaporator vessel which operates under a partial vacuum produced by two steam ejectors in series. The liquor at approximately 60°C, is heated by steam pokers which penetrate the roof of the vessel. These pokers can be individually replaced, if required, unlike the more conventional heating coils which are virtually impossible to replace. The steam condensate, along with the evaporator overheads and the vacuum ejector exhaust, are monitored and conducted via the LA drain for sampling, sentencing, filtration and disposal. The salt concentrate is ejected on a batch basis to a storage facility to await treatment in EARP.

A vessel ventilation system is provided to ensure containment of any vapours issuing from the process liquors. The extract air, and vapours from the plant vessels, are drawn through a recirculating caustic scrubber system. Caustic scrubbing is used to increase the removal of radioactive iodine which is present in small amounts in the feed liquors. Downstream of the scrubber the gases pass through primary and secondary HEPA filters before being discharged via an existing stack system. The scrubber produces a low volume caustic effluent which is monitored and sent to the sea discharge sentencing system.

Figure 6 shows a plan of the Salt Evaporator building. When fully operational, at a cost of about $22M, the plant will provide two identical cells. The A cell is complete and was commissioned in March 1985, and will provide the normal processing route. The B cell, when completed in early 1988, will act as a backup and will permit flexible operation since the process can be diverted from one cell to the other at various stages. Each line will be capable of processing 150m^3/day of feed liquor and all services to the plant have been sized on this basis.

The Enhanced Actinide Removal Plant (EARP)

Medium active and salt evaporator concentrates are currently stored. EARP is being constructed, at a cost of $300M, in order to extract alpha, and some beta activity from the comparatively low volumes (about 1000m^3/year) of these concentrates and the higher volumes (about 250m^3/day) of low active effluents.

Figure 7 shows a simplified process flow diagram for EARP. Both feeds to the plant result directly from reprocessing operations and are, therefore, acidic and contain significant amounts of iron in solution (up to 40 tonnes/year in the high volume stream). A study of the methods available for the treatment of both EARP feeds has shown that flocculation, produced by the addition of sodium hydroxide to the iron-bearing acidic streams to increase the pH from 0.5 to 9.5, is the most effective process as almost all of the alpha activity co-precipitates with the floc, leaving a virtually inactive aqueous phase. Development work, which is continuing, has indicated that by the addition of relatively small volumes of specific chemicals it is possible to improve the beta activity removal, particularly in the case of the concentrates.

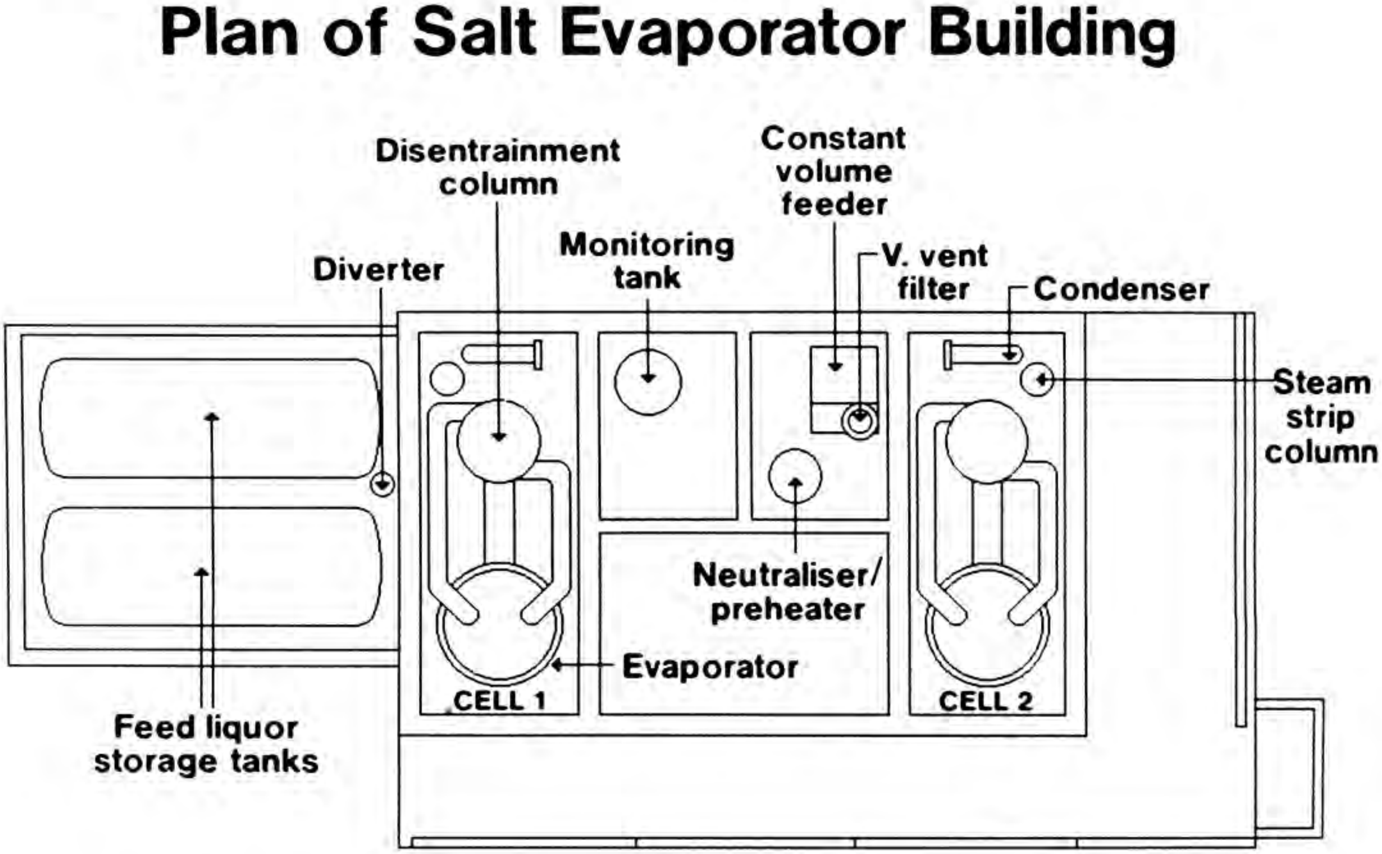

Fig. 6. Plan of Salt Evaporator Building.

BNFL

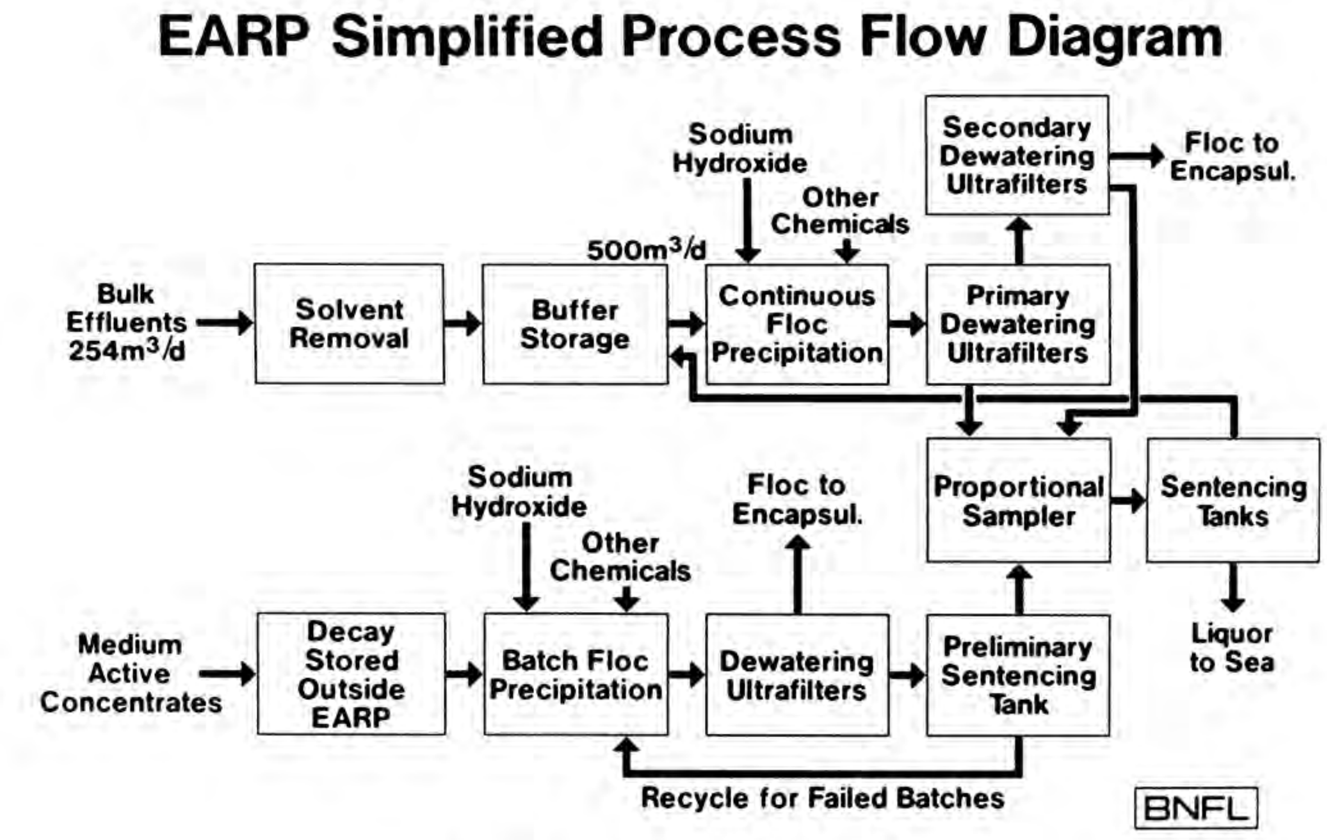

Fig. 7. EARP Simplified Block Diagram.

Development work has also shown that higher decontamination factors can be obtained if the low volume concentrate streams are treated separately on a batch basis. The bulk (low active) effluents are treated on a continuous basis in a separate line.

The active ferric floc must be separated from the liquor to give a solid waste suitable for encapsulation in cement and this will be done by ultrafiltration. The aqueous permeate is discharged to sea after sentencing on a batch basis to ensure that it meets the discharge criteria. Any batches which fail to meet the set requirements are recycled within the plant for further treatment.

The satisfactory operation of the ferric floc process is dependent on the level of iron in the feed to the plant. Iron will be added to the feed to bring it up to the minimum level required to achieve an acceptable decontamination although, during operation of the fuel reprocessing plants, the amount of iron in the effluents being fed to EARP is generally adequate to meet the process requirements. Because it arises in discrete batches rather than on a continuous basis, provision has been made in EARP to smooth out the peaks by feeding via a buffer tank having a capacity equal to one day's feed. During reprocessing plant outages, iron will be added to the other EARP feeds, as necessary, to achieve the required minimum levels.

In order to meet the required site discharge targets, all the effluent produced by the selected feed sources must be treated in EARP. It is, however, recognised that, like any other process plant, EARP will be subject to short term maintenance and breakdown outages. To cope with any such outages, EARP has been provided with sufficient buffer storage capacity for three days volume of feed arisings. This buffer store has been combined with the feed buffer to smooth out variations in iron feed level.

Hence to clear backlogs arising as a result of EARP outages, and the need to recycle any batches of treated effluent which, when sentenced, do not meet the discharge criterion, the main process sections of EARP have been sized to treat double the normal anticipated feeds ie $500m^3$/day of bulk effluents and around $2400m^3$/year of concentrates.

The two major requirements of the solid/liquid separation stage are a high separation efficiency and the production of a solid waste which has been adequately dewatered to allow its direct encapsulation in a cement-based matrix.

The principle of ultrafiltration requires that the liquid to be filtered is pumped, at pressure and at relatively high velocity, typically about 5 m/s, through a tubular membrane. The high velocity reduces the build up of solids on the membrane wall (surface filtration), whilst the pressure induces a flow of permeate through the membrane wall (cross flow filtration). The pore size of the membrane coating is typically a few hundredths of a micron and is often expressed in the molecular size of materials they will filter. A recirculation loop through the tube is kept pressurized by pumping new feed to be filtered into the circuit at the same rate as the permeate flows out of the tube. The flow of permeate through a single tube is, of course, small and hence ultrafiltration modules are built up of many tubes in parallel, similar to a shell and tube heat exchanger.

Although surface filtration can be minimised, development work has been carried out to identify techniques for backwashing and chemical cleaning. The floc produced by the EARP process can be dissolved in nitric acid and this, therefore, gives a high efficiency method of cleaning fouled tubes. Despite this, it is accepted that ultrafilter membranes will have a finite life. Wear of the coating, or mechanical damage to the graphite tubes, could also result in the need to replace membranes. Work carried out to date on the use of ultrafilters for floc dewatering, and investigation of their performance in other industries, has indicated that a life of at least three years can be expected before replacement is necessary. The standard ultrafilter units used in non-nuclear industries do not lend themselves to remote replacement within very low radiation uptake limits. BNFL has therefore developed and tested a remotely maintainable cartridge design based on the principles used for the SIXEP pumps and valves.

The initial floc as produced by the EARP process contains only a few hundreds of ppm of ferric hydroxide and is a liquor very closely resembling water in physical properties. The dewatered floc in the form required for encapsulation in cement contains up to 100 grams per liter of iron, as ferric hydroxide, and is a thixotropic material with viscosity of about 7 poise. Development work has shown that this material can be handled by the ultrafilters and that it can also be pumped. Dewatering of flocs from the bulk effluents in EARP will, therefore, be carried out in two stages. In the first stage, dewatering will be carried out to a level of 5-10 grams per liter of iron as ferric hyroxide. At this concentration the liquor is still very similar to water in physical properties and can be handled by conventional pumps. In the second stage, final dewatering to up to 100 grams per liter of ferric hydroxide is carried out. Since up to ninety per cent of the water is rejected in the primary ultrafilters, the scale of the secondary units, which handle the more viscous materials, is substantially reduced. In the case of the much smaller volume of liquor produced from the treatment of 25m^3 batches of concentrate, dewatering is carried out in a single stage.

Figure 8 shows a plan of the EARP building. The receipt tanks can be seen at the bottom left, the neutralization vessels, in which floc precipitation occurs, at the top centre, the ultrafilters in the middle left and the sentencing tanks at the bottom right. The building concept was intended not only to provide adequate maintenance access but also to maximise modularization of plant and pipework.

CONCLUSIONS

The three plants described above, when they are all fully operational, have and will contribute significantly towards making the reductions in activity discharges shown in Figs. 1 and 2. However, they are only part of a comprehensive

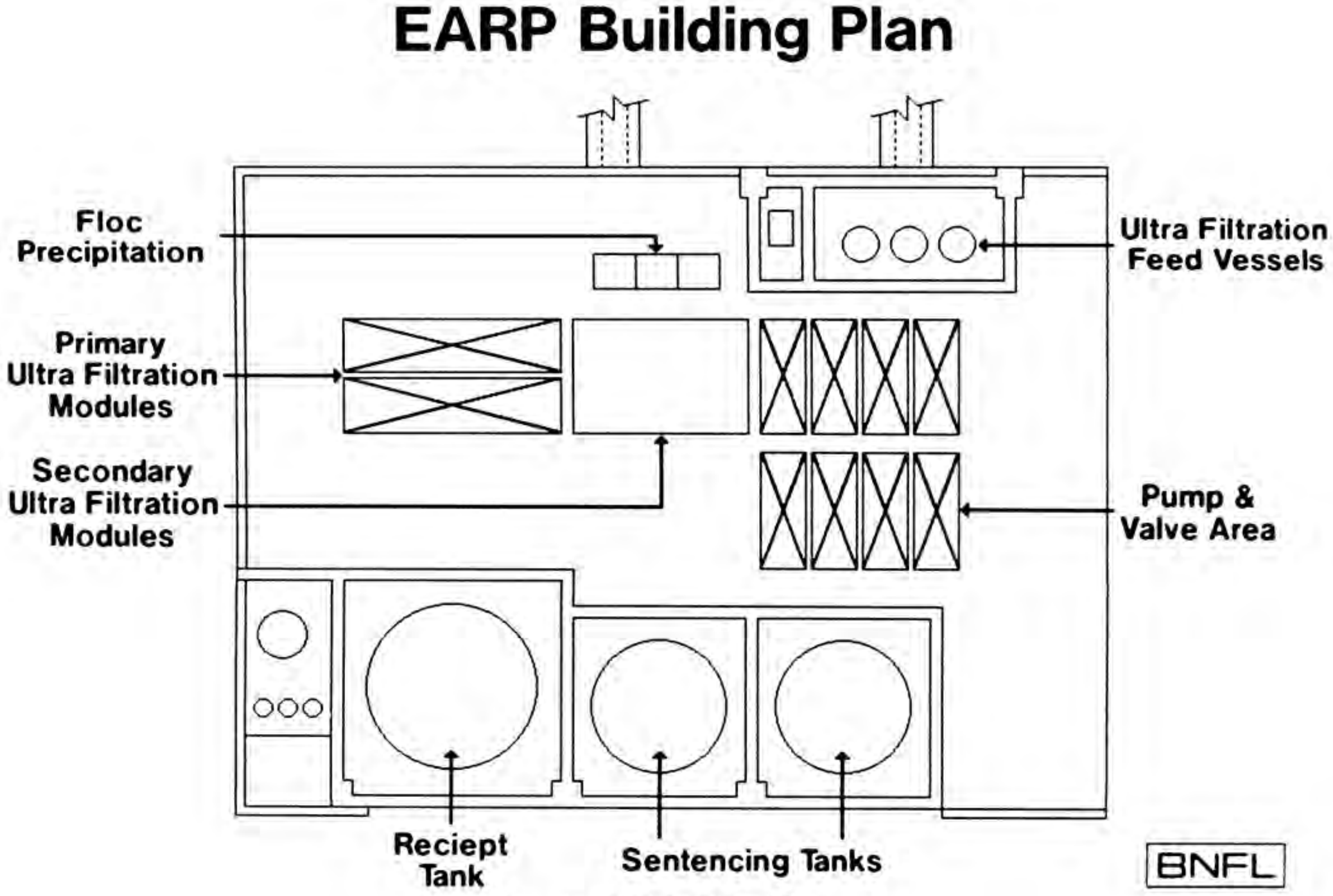

Fig. 8. EARP Building Plan.

overall effluent management philosophy which is currently being developed for the Sellafield site. Other measures to be taken include:

1) The refurbishment of ageing plants to the higher standards now required.
2) Increased segregation of selected streams to enable treatment plants to be used most efficiently.
3) Removal of solids and solvent from effluent streams.
4) Sentencing of streams prior to discharge.

Implementation of this philosophy requires substantial investment in capital and resources. Nevertheless the Company has to maintain its competitive position in the fuel cycle services business and to this end every effort is being made to minimise capital, operating and maintenance costs by the use of modular designs, remote handling techniques, common services and other features.

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