

MANAGEMENT OF WASTES FROM THE REFINING AND CONVERSION OF URANIUM ORE CONCENTRATE TO URANIUM HEXAFLUORIDE

REPORT OF AN ADVISORY GROUP MEETING
ON WASTE MANAGEMENT
OF URANIUM REFINING FACILITIES
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
AND HELD IN VIENNA
17-21 DECEMBER 1979



A TECHNICAL DOCUMENT ISSUED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1981

**MANAGEMENT OF WASTES FROM THE REFINING AND CONVERSION
OF URANIUM ORE CONCENTRATES TO URANIUM HEXAFLUORIDE
IAEA, VIENNA, 1981**

Printed by the IAEA in Austria
February 1981

**PLEASE BE AWARE THAT
ALL OF THE MISSING PAGES IN THIS DOCUMENT
WERE ORIGINALLY BLANK**

The IAEA does not maintain stocks of reports in this series. However, microfiche copies of these reports can be obtained from

**INIS Microfiche Clearinghouse
International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400 Vienna, Austria**

on prepayment of US \$1.00 or against one IAEA microfiche service coupon.

FOREWORD

Non-radioactive effluents from the nuclear industry are today small on an absolute basis and miniscule compared to other important industries and are not specific to the nuclear industry. Adequate technology for treating and monitoring effluents exists so that the future growth of the nuclear industry should pose no significant non-radiological pollutant problem either on a national or global basis.

The minimization of environmental effects from non-radioactive pollutants in the nuclear industry can be achieved by the choice of process flow sheet for each step of nuclear fuel cycle.

This report is the outcome of an IAEA Advisory Group Meeting on "Waste Management Aspects in Relation to the Refining of Uranium Ore Concentrates and their Conversion to Uranium Hexafluoride", which was held in Vienna from 17 to 21 December 1979. The report summarizes the main topics discussed at the meeting and gives an overview of uranium refining processes, being used in nuclear industry. The meeting was organized by the International Atomic Energy Agency, Radioactive Waste Management Section. Mr. Y. Zabaluev was the Scientific Secretary of the meeting.

The IAEA wishes to express its thanks to all the participants who contributed to the preparation of this report. The initial draft of this report was prepared by Mr. J.F. Wing (USA) and Mr. M.R. Hayes (UK). The Agency wishes, in particular, to express its gratitude to Mr. J.F. Wing who served as Chairman of the meeting and contributed considerably to the editing of the material.

TABLE OF CONTENTS

1. Introduction	7
2. Environmental considerations affecting siting of the facility and discharge limits	9
3. Waste requiring management and treatment of these wastes	14
4. Maintenance and Monitoring	27
5. Decommissioning	29
6. Responsibilities	32
References	35
Bibliography	36
Appendix I – Flow Sheets	37
Appendix II	63
Definitions of terms used in the report	67
List of participants	71

1. INTRODUCTION

Commercial production of uranium hexafluoride (UF_6) began in the mid-1950's. As of 1980 there are about ten uranium refineries in nine of the industrialized nations, converting uranium concentrates containing 60-80% uranium to uranium hexafluoride. With the expanding use of nuclear power the need for additional uranium refining capacity is recognized. There are several developing nations that have a significant number of identified uranium ore deposits which will eventually be mined and processed. Since most of these deposits are quite remote from the existing uranium refineries, it could be advantageous to refine the concentrate closer to the mines and concentrate mills. These thoughts have led to the issuing of this Technical Report on the management of wastes and effluents associated with uranium refining.

This report describes current practice at the time of writing and makes recommendations as to the best practicable technology for the management of refinery wastes. The two guiding principles are, 1) to minimize the quantity of waste requiring management, and 2) to minimize the environmental impact of those wastes which cannot be eliminated. While this document is directed toward management of effluents and wastes associated with uranium refining, it is recognized that there are other areas which require attention such as health protection and environmental protection. Although this Technical Report does not address these latter areas in detail, the bibliography includes appropriate references in these areas.

In the early planning stages there are at least two financial aspects which need to be carefully considered. In the first case, it may not be unusual for the competent authority to require the owner/operator to post a performance bond which could represent a substantial sum. The purpose of this bond would be to assure responsible conformance with applicable regulatory rules and conditions. In the event of serious failure, the bond would cover the cost of such remedial actions as may be necessary. Another important financial aspect which should be carefully considered is the establishment of a fund through a financial plan to accommodate the cost at final refinery closure of eventually decontaminating, decommissioning, and/or dismantling the facility. Although this cost may be highly speculative and the specific requirement for such actions would most likely be determined by the competent authority, it should be included in an initial comprehensive financial plan and adjusted periodically, as needed.

2. Environmental considerations affecting siting of the facility and discharge limits

The environment at the site of the uranium refinery has a very great influence on the requirements for successful waste management. The major site characteristic which affect the selection of waste retention sites and waste management practices are climate, the magnitude and frequency of floods, geography, topography, demography, geology, seismicity, hydrology, flora and fauna and the revegetation potential.

Authorized limits for discharge of effluents should be fixed in each case by the competent authority, which will take into account the radioactive and chemical components of the effluent, the characteristics of the environment, and the ICRP recommendations, which aim to limit radiation exposure of the critical group of the concerned population to values below the fixed limits and to optimize protection, maintaining all exposures due to the release as low as is reasonably achievable, economic and social considerations being taken into account.

The aim of environmental monitoring should be to check whether approved procedures and regulations have been followed, to ensure that nothing unforeseen has happened, to ascertain that individuals are not and are not likely to be exposed to contaminants in excess of approved limits and to indicate whether discharge authorizations should be redefined.

A site should be selected so that the existence of any residual activity after decommissioning will not significantly change the local environment.

2.1. Climate

Precipitation, especially when considered with the annual evaporation rate at a site, virtually determines whether all the liquid wastes can be retained in waste retention ponds [1] or whether liquid waste will have to be discharged to the environment. If the average annual evaporation rate exceeds the average annual precipitation rate, and assuming zero or estimated seepage losses, it is relatively simple to calculate the area of the waste retention ponds required to evaporate all the effluent. Consideration must then be given to the distribution of the rainfall, both throughout the year and from year to year, to ensure that adequate freeboard in the waste retention ponds is available at all times. For such ponds it is essential that the catchment area draining to the ponds is not much greater than that of the pond system itself. If rainfall is unevenly distributed, it may mean that rivers or streams will dry up during the dry season, so that effluents cannot be discharged to them. Another effect which has to be considered is the possible seasonal variation in the water table, which could affect the rate of seepage from the waste retention system [1] and the rate of transport of contaminants through any aquifer which may be present. In some situations an impervious liner may be required for the waste retention ponds.

The magnitude and frequency of floods must be considered when siting a waste retention system, as it is generally unwise to locate a system on a site that may periodically be subject to flooding. The diurnal and seasonal fluctuations in surface ground temperatures also influence waste management. Severe freezing and thawing conditions can present serious problems to shallow land burial due to surface heaving, settling, etc.

The atmospheric inversion characteristics of the facility site are an important parameter in considering discharges to the atmosphere. A knowledge of the frequency, duration, strength and break-up interval for atmospheric inversions is needed for the calculation of the concentration of airborne contaminants both within and external to the site, as is a knowledge of local wind speed and direction.

2.2. Geography

This factor also has a large influence on the waste management practices adopted at a particular site. For instance, the topography may not allow the construction of large impoundments or permanent rivers may not be available to dilute effluent discharges. Adjacent farms, towns, or cities may impose additional constraints. The present and potential land use for terrestrial and aquatic food production determines the dose commitment that would result from the discharge of a given quantity of radioactive material. The age, diet, recreational and working habits of the local inhabitants determine the characteristics of the critical group used for radiological calculations.

2.3. Hydrology

The hydrology of the area influences the amount of water infiltrating into the site and in some cases into the waste retention system, and controls the length of time that the water is in contact with the wastes.

Where suitable hydrogeological conditions can be demonstrated to exist, it may be satisfactory to dispose of effluent by injecting it into a confined underground aquifer containing non-potable water. This decision is affected by the existing and potential uses of ground-water in the area. Competent authority approval would be required.

Discharge of liquid wastes into streams or rivers may be acceptable if dilution factors can ensure sufficiently low levels of pollutants and if existing water quality is sufficiently good that the incremental loading due to the refinery will not overstress the local ecology. Particular care should be taken to avoid discharge or potential discharge into small surface streams, where the waste volume may represent more than a few per cent at the stream flow. Usually more stringent discharge limits are applied to effluents entering small streams since they have less assimilative capability.

2.4. Flora and fauna

The nature of aquatic and terrestrial habitats influences the degree of environmental degradation that would result from inappropriate waste management. Siltation can cause loss of fish breeding grounds and food as well as reduce egg hatchability. Discharged acidity affects plant nutrition and in severe cases leads to loss of bank vegetation and bank stability. Fish and zooplankton are very sensitive to heavy metal ions and are more sensitive, the softer the water. Thus fairly minimal waste discharges can upset the local ecosystems and produce substantial changes in the diversity of species. This in turn can reduce the commercial and recreational value of the waterbody.

Water quality also influences the bioaccumulation factor of aquatic organisms for radioactive and non-radioactive heavy metals. These factors need to be taken into account when assessing the radiological impact for members of the critical group. The soil-pasture-stock transfer mechanism determines the capacity of the terrestrial environment for heavy metal wastes.

The presence of contaminants such as nitrates, ammonia and fluorides in the liquid effluent may present a significant environmental threat if not adequately treated. Fluorides can damage vegetation, especially sensitive species as well as cause fluorosis in grazing animals. Ammonia can produce fish kills and supply excessive nutrients to the aquatic system. Nitrates can also be a source of excessive nutrients to the aquatic system as well as be a source of drinking water contamination. The competent authority should be consulted early in the planning stage for advice on discharge limits for both airborne and liquid effluents.

3. Wastes Requiring Management and Treatment of these Wastes

This section discusses those wastes from uranium refining and conversion operations, which could present a detrimental impact to man and the environment and thus require management in accordance with the authorisations granted by a competent authority. In most instances where it is required to reduce the level of a contaminant in a waste stream, it is most cost effective to confine and treat the waste stream as near its source as is reasonably possible. At such a point the volume will be near its lowest and the concentration near its highest, resulting in the opportunity for a smaller, more efficient, treatment process.

Standard practice for the production of uranium ore concentrate employs milling and leaching of uranium bearing rocks or shales followed by separation of the uranium using either solvent extraction or ion exchange techniques with final precipitation as an ammonium, alkali or alkaline earth diuranate. During this process the uranium is separated from the majority of associated radionuclides and other impurities. The diuranate is then dried and/or calcined prior to drumming for shipment. Flow sheets of the typical refining processes are shown in the Appendix 1.

Other than the uranium the only radioactivity of any importance associated with uranium chemical concentrate arises from the short half-life daughter products ^{234}Th (UX I) and ^{234}Pa (UX II) together with minor traces of ^{230}Th , ^{226}Ra and daughters and of natural thorium (^{232}Th). Other impurities are largely any remaining ammonium or alkaline earth cations sulphate, phosphate, fluoride and metal impurities at low to trace levels. These relatively low levels of radioactivity, together with any uranium lost during the refining operation, other impurities

in the uranium concentrate and reagents used during refining, give rise to the wastes which may escape to, or are discharged to, the environment.

There are a number of alternative processes for the refining and conversion of uranium concentrate to the volatile uranium hexafluoride of a quality suitable for feed to a uranium enrichment process. However only two, the dry hydrofluor and the wet solvent extraction route, are widely used or envisaged for the foreseeable future. Although further development of the technology may be expected to lead to modifications to the plant and equipment for the different stages of these processes, no significant changes in waste arisings are foreseen. Wastes arising at each stage of the refining and conversion process are considered. These considerations will apply, as appropriate, to both the wet and dry processes.

Basically the dry hydrofluor process eliminates impurities either as volatile compounds or as solid constituents of ash. The wet solvent extraction method separates impurities by extracting the uranium values in an organic solvent leaving other constituents in the aqueous phase.

The dry hydrofluor process consists of the following operations:

- (a) Pre-process handling, weighing, sampling and storage;
- (b) Fluidized-bed reduction of the U_3O_8 with cracked ammonia to UO_2 ;
- (c) Fluidized-bed hydrofluorination with anhydrous HF to a crude UF_4 ;
- (d) Fluidized-bed fluorination with elemental fluorine to crude UF_6 ;
- (e) Fractional distillation to refined UF_6 .

In the last stage of the process the impurities are separated from the product by taking advantage of relative differences in volatility.

The second process is a wet chemical solvent extraction method followed by denitration, reduction, hydrofluorination and fluorination in fluidized bed reactors. The method consists of the following operations:

- (a) Pre-process handling, weighing, sampling and storage;
- (b) Digestion in hot nitric acid;
- (c) Counter-current solvent extraction with tributyl phosphate in hexane, dodecane or kerosene;
- (d) Re-extraction of uranium as uranyl nitrate solution;
- (e) Calcining to UO_3 ;
- (f) Fluidized-bed reduction with hydrogen or cracked ammonia to UO_2 ;
- (g) Hydrofluorination in a countercurrent reactor to UF_4 using anhydrous HF;
- (h) Fluorination to UF_6 by reaction with elemental fluorine in flame reactors or fluidized beds.

Block flow diagrams of the two basic processes are shown in Appendix I. Also included are the flow diagrams describing the process in use in France, England, Canada and the United States.

3.1. Feed Materials

Uranium concentrate, the most common feed material, may consist primarily of ammonium diuranate $(NH_4)_2U_2O_7$ or triuranium octaoxide (U_3O_8), uranium trioxide (UO_3), ammonium, alkali metal or alkaline earth uranates

(XU_2O_7) or a variable mixture of all of these. Typically, the composition of radionuclides in the concentrate may be :

Uranium (as U)	60-80%	
Ionium (^{230}Th)	3×10^3 - 1×10^4	pCi/g U
Radium (^{226}Ra)	20-400	pCi/g U
Thorium (^{232}Th)	100-2000	pCi/g U

Short half-life daughters of the uranium (^{234}Th , ^{234}Pa) and of radium and thorium can normally be assumed to be present in equilibrium amounts. Separation of uranium daughters can occur at more than one stage in the process as a result of ingrowth of the ^{234}Th , (half life 24.1 days).

Impurities in uranium concentrate may include S, SO_4^{2-} , PO_4^{3-} , CO_3^{2-} , F^- , Cl^- and organic matter, together with trace metal impurities such as, Al, As, B, Ba, Bi, Ca, Co, Cu, Fe, K, Mn, Mo, Mg, Na, Ni, Nb, Pb, Sb, Si, Ta, Ti, V, W, Zr and rare earths.

Other possible feed materials include impure uranium concentrate with a high fluoride content recovered from the phosphate industry and relatively pure uranium slurries obtained by heap-leaching and in-situ mining operations.

3.2. Pre-process handling, weighing, sampling and storage

Handling and storage of uranium concentrate should preferably be carried out in an area from which any spillages or washings can be collected, for example in a sump or settling pit, so that uranium losses can be returned to the process.

Typical radiation levels from stacks of drummed uranium concentrate are of the order of 2-5 mr/hr. so that stacking close to perimeter fences should be avoided.

The opening of drums for inspection, weighing and sampling together with re-closure, should be carried out in a total enclosure with air extraction leading to primary and secondary air filtration prior to discharge to atmosphere via a stack.

Losses of uranium to the air filtration system are shown in Appendix II. Dust collected in the primary filter is returned to the process. In some cases disposal of secondary filters by incineration followed by recovery of the uranium may be possible and is to be encouraged. Alternatively these may be disposed of by burial. These general considerations of containment, air extraction and air filtration may be taken to apply to all stages of the processes handling solid uranium compounds.

3.3. Feeding

The drum opening and emptying operations should be carried out in an enclosure connected to a ventilation system. At a throughput of 10,000 t U/yr some 40,000 empty drums will arise. Depending on the economics, it may be possible to clean and recondition the drums for reuse within a closed cycle between the concentrate supply and the refinery. Alternatively, facilities for washing and disposal of these by crushing and burial may be provided. A further alternative may be controlled smelting of the contaminated drums with return of the uranium bearing dross to the process.

3.4. Dissolution and Digestion in Hot Nitric Acid

Dissolution and digestion is normally carried out in a multiple-effect dissolver using hot nitric acid. The process generates a mixture of oxides of nitrogen (NOx) and acid fumes which should be passed to a nitric acid recovery system.

The amount of nitric acid required for dissolution will vary according to the type of concentrate, as will the amount of NOx arising.

Dissolution may result in the release of a small quantity of radon ($^{222}_{\text{Rn}}$ approximately 0.5 Ci/yr) which is released to atmosphere via the nitric acid recovery system.

Liquor from the dissolution stage may be filtered to remove silicious solids which contain entrained uranium, traces of radium and thorium isotopes. These solids are normally reslurried and refiltered to recover the uranium prior to disposal by shallow land burial.

In some cases addition of phosphate or fluoride ions is made at the dissolution stage to precipitate or control specific impurities. In such cases the uranium liquor is fed to the solvent extraction stage without filtration

3.5. Liquid-liquid extraction of uranium

The uranium is extracted from the feed solution or slurry in a counter-current liquid-liquid extraction system, using tri-n-butyl phosphate in kerosene, hexane or dodecane. This produces an aqueous raffinate stream which contains trace amounts of uranium, uranium

daughter activities, all the other radionuclides and trace metals from the feed as a mixture of nitrate salts, and 1 to 3N free nitric acid. The uranium is re-extracted into the aqueous phase by counter-current contacting of the loaded solvent with warm dilute nitric acid solution to give a pure uranyl nitrate solution.

There are a number of options for treatment and disposal of the raffinate.

- (a) Nitric acid may be removed by adding sulphuric acid followed by evaporation. The resultant slurry may be returned to a uranium mill for recovery of the uranium, in which case the remaining wastes would be handled in the mill tailings system.
- (b) A denitration process similar to that used for the uranyl nitrate can also be employed to obtain a dry virtually nitrate-free, solid for disposal. An ammonia-free feed would be required to use this option.
- (c) The raffinate can be treated chemically to precipitate the radionuclides and heavy metals, which are commonly separated in a settling pond. The resultant clean nitrate solution may be recycled in the chemical industry for such uses as fertilizer. In certain situations in the U.S. the solution has been applied directly to pasture areas for fertilizer 2 .
- (d) Biological denitrification may also be used as a method of destroying the nitrogen compounds in the liquid, but this requires a source of organic carbon and favourable weather conditions [3-8].
- (e) In a climate with positive evapo-transpiration, the raffinate may be discharged to ponds and allowed to evaporate.

- (f) In some circumstances, disposal of the liquid waste to the ocean may be acceptable.

Options, such as settling ponds, which result in residual radioactivity after the bulk of the wastes have been evaporated, recycled, etc., will require provisions for final disposal when the refinery ceases operation. In some circumstances it may be possible to convert the waste storage into a permanent repository. For other cases, the wastes will have to be removed to a waste disposal area. In recognition of this possibility, it is prudent to minimize the waste volumes and any dispersal of the waste during the refinery operations.

Gradual degradation of the tri-n-butyl phosphate/diluent occurs during the solvent extraction cycle. This is rectified by periodic chemical treatment of the solvent using sodium carbonate solution. In the event that the solvent can no longer be returned to a satisfactory condition by this treatment disposal becomes necessary. The preferred method of disposal would be by incineration under controlled conditions.

3.6. Evaporation

Evaporation of the pure uranyl nitrate to a highly concentrated solution is carried out in a closed multiple effect evaporator designed such that nitric acid vapour and any entrained uranium are returned to the nitric acid recovery system and thence to the main process stream.

3.7. Ammonium Diuranate Precipitation

In some processes the uranium is recovered from the pure uranyl nitrate solution by precipitation with ammonia and filtration

of the resulting ammonium diuranate. The filtrate and washings from this process contain ammonium nitrate but low radioactivity. As such, they may be disposed of as a saleable by-product on the commercial market following additional treatment.

3.8. Denitration

Thermal denitration of concentrated uranyl nitrate liquor, is carried out with continuous agitation in various types of equipment. The denitrator should always be connected to a system for recovery of nitric acid and entrained uranium prior to discharge of the residual gases to atmosphere via a stack. A further reduction in the emission of oxides of nitrogen may be achieved using catalytic conversion.

3.9. Drying and Calcining of Ammonium Diuranate

Drying and calcining of ammonium diuranate releases moisture, ammonia and traces of ammonium nitrate. These are removed in a wet scrubber, the liquor from which is low in radioactivity and sufficiently pure for sale on the commercial chemical market. Careful monitoring of this material is recommended, and in some cases, additional treatment may be necessary before it can be unconditionally released.

3.10. Reduction of Uranium Oxides to Uranium Dioxide

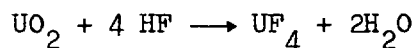
Reduction of higher oxides of uranium is carried out using fluidised bed, moving bed or kiln equipment. In any case the reducing gas stream, hydrogen or cracked ammonia, is diluted with nitrogen and the off gas from the process is passed through either sintered metal filters or a wet scrubber system to remove entrained uranium.

Some of the sulphate contained in the uranium oxide is reduced at this stage and may pass to atmosphere as hydrogen sulphide. Alternatively the hydrogen off-gas may be burned, in which case the hydrogen sulphide is oxidised to sulphur dioxide prior to discharge.

3.11. Hydrofluorination

Hydrofluorination is carried out using fluidised bed, moving bed or kiln equipment and hydrofluoric acid as reagent.

According to the reaction:



unreacted hydrogen fluoride gas will result in dilute hydrofluoric acid after condensation or wet scrubbing of the off-gas stream. Therefore, although the hydrofluoric acid is recovered, it is not in a suitable form for return to the process and is normally either collected for sale or treated with lime or ammonia followed by lime. The resulting calcium fluoride may be disposed of to the commercial market with proper monitoring or by burial.

Any remaining sulphur compounds in the uranium dioxide also appear in the off-gas from the hydrofluorination stage as hydrogen sulphide. Where condensation of hydrofluoric acid from the off-gas stream is used, the residual gas may be scrubbed to remove hydrogen sulphide and any remaining hydrofluoric acid. Depending upon the process utilized monosodium sulphide will be a component in the liquid effluent.

12. Fluorination to Uranium Hexafluoride

Fluorine is produced as required by electrolysis of hydrofluoric acid in molten potassium hydrogen fluoride (KF.2HF) so that, other than minor losses in the process, no problems associated with fluorine storage occur.

Residual electrolyte from the electrolytic cells is dissolved in water and the resulting liquor subjected to lime treatment followed by separation of the calcium fluoride.

Removal of hydrogen fluoride from the fluorine process gas stream may be by condensation at low temperature (-80°C) or by the use of sodium fluoride. In the latter case the spent sodium fluoride may be regenerated or disposed of by burial.

Hydrogen from the electrolytic cells may be used in the facility, scrubbed to remove hydrogen fluoride and discharged to atmosphere, or burned in a system designed to recover traces of fluorine and uranium hexafluoride. In the latter case the resulting gases pass to a scrubber system.

If the fluorination is carried out in a fluidised bed process, it is necessary to provide a bed of calcium fluoride to ensure the correct mode of fluidisation. Trace metal impurities, uranium daughter products and non-volatile metal fluorides accumulate on the calcium fluoride so that the calcium fluoride itself and any solids carried over to the off-gas filtration system are relatively radioactive. Disposal of the spent calcium fluoride may be by land burial. The material may be stored to allow decay of the uranium daughter product activity for ease of handling. A storage period of six months will result in the decay of ^{234}Th to about 0.5% of its original value.

Uranium hexafluoride is desublimed from the filtered reactor off-gas and the residual fluorine may be recycled to the reactor or passed to a secondary clean-up reactor. Any remaining unreacted fluorine is then removed in a scrubber system prior to discharge of the gases to atmosphere.

In the case of a flame reactor an ash is formed consisting of unreacted UF_4 , UF_5 and other metal fluorides and containing the radioactive uranium daughter products. This ash is recycled to the fluorination process or to the dissolution stage.

3.13. Distillation of Uranium Hexafluoride

In the dry process, purification of the uranium hexafluoride produced in the previous step is accomplished by fractional distillation. Impurities removed from the UF_6 during distillation are normally recycled to other phases of the process.

3.14. Cylinder Filling

Product uranium hexafluoride is liquified prior to filling transport containers.

3.15. Recovery of residues

Residue arising from cleaning, and decontamination, scrubber residues, ash and the like are acid leached to dissolve the uranium and the resulting solution is returned to the main process, prior to the solvent extraction stage. Calcining and a preliminary separation of uranium as ammonium diuranate may be used in some recovery processes.

In the dry process residues are treated by a wet carbonate leach and the extracted uranium is returned to the process.

In all these recovery operations the wastes arising are combined with those from the main process and treated as described in the previous sections.

Where caustic scrubbing solutions are used for off-gas treatment, these may be treated chemically to remove uranium and to precipitate fluorides as calcium fluoride. The uranium can be recycled to feed dissolution and the calcium fluoride can be disposed of as indicated in Section 3.11.

3.16. Additional waste arisings

It is assumed that a facility will be equipped with analytical laboratories, incinerator, laundry and sewage treatment. Estimates of the wastes arising from these are shown in Appendix II for airborne, solid and liquid wastes and for materials arising from routine maintenance and refurbishing operations.

These wastes are treated as described previously for the recovery of uranium.

4. Maintenance and Monitoring

4.1. Maintenance

Routine inspection and maintenance of process equipment and effluent treatment facilities should be carried out to ensure efficient plant operation and therefore minimal losses of pollutants to the environment.

4.2. Monitoring

4.2.1. Monitoring must detect both radioactive and chemical pollutants.

Effective environmental monitoring requires that a comprehensive assessment of the environment be made prior to construction of the facility. This should include sufficient understanding of the ecology of the area for early warning of adverse effects on the environment to be obtained.

4.2.2. Process - Monitoring and sampling of process streams should be systematically carried out to prevent malfunctions which could give rise to additional and/or increased effluent discharges.

4.2.3. Solid Wastes - To ensure that solid wastes are under control, it is clearly necessary that they be monitored from time to time. Where the environment may be affected, appropriate samples must be taken and examined to measure the effect. This is in addition to samples which may be taken to comply with discharge limitations.

4.2.4. Airborne Wastes - Stacks through which off-gases from the various processes are emitted should be equipped with monitors

to measure the particulate and gaseous materials discharged from these processes. In addition, deposition collectors should be strategically positioned within and outside the plant perimeter. These samples should be analysed routinely to back up the results of the stack monitors.

4.2.5. Liquid Wastes - Water samples from effluent discharge arrangements, settling ponds, lagoons and storm water drains should be routinely analysed.

4.2.6. Environment - Environmental samples selected to detect damage to the environment, may include such materials as stream and aquifer water, fish, stream sediments, animals and agricultural products.

4.3. Responsibilities - The manager should be responsible for ensuring that such inspections and monitoring, including sampling and analysis to determine that approved waste management practices are being followed, are carried out during operations and until the site is decommissioned. The owner should be responsible after that period. This ensures that there is a clear and unambiguous responsibility at all times. See Section 6.2.

5. Decommissioning

- 5.1 Design. Any nuclear plant should be designed in such a way as to enable decommissioning to be carried out with maximum safety and ease. This consideration also applies to any waste accumulated on the site, during the operational life of the plant or at the time of its decommissioning.
- 5.2 Contaminated plant and structural material. All items of plant, equipment and structural material contaminated with radioactive material should be decontaminated to or below the approved level before they are removed from the site. Alternatively, they may become another type of solid waste and should be disposed of in accordance with approved procedures. Smelting should be considered as a final means of metal decontamination with the dross being buried at an approved location.[9-13].
- 5.3 Contaminated Land areas. All areas contaminated with radioactive material should be decontaminated to the approved level. Soil scrapings can be returned to a uranium mill for processing or buried in an approved location.
- 5.4 Waste retention systems. Waste retention systems should be left in a completely stable form so that they blend aesthetically with the general landscape. Appropriate measures should be taken to protect the public from any hazard presented by the waste retention systems. The competent authority may require stabilisation measures such as capping drains, filling in retention ponds, mounding to approximately one metre above the pond centre, and revegetating the slope.

5.5. Site. Actions taken to decommission the site should ensure that releases of radioactive and toxic material are within approved limits and minimize radiation exposures. It is expected that after decommissioning some land may be subject to a covenant restricting future use of the land. The land areas for which restrictions are necessary should be minimized to the extent practicable by the use of proper practices during operation and decommissioning.

Particular importance will be attached to the restrictions imposed by wastes contained in the "waste retention system". In an area where natural radioactivity is already elevated, a waste storage system could be converted to a waste disposal system during decommissioning, whereas, in areas of low natural radioactivity this might not be acceptable.

The choice of a site should be based on a cost-benefit analysis, comparing criteria dictated by ease of operation with criteria dictated by ease of decommissioning.

5.6 Financial arrangements for maintenance and monitoring of facilities after decommissioning. The owner/operator should establish approved financial arrangements (e.g. bonding, escrow accounts) to ensure that there are sufficient funds to carry out the maintenance and monitoring programmes.

5.7 Periodic inspections of wastes and the environment after decommissioning. The owner of the decommissioned site should ensure that inspections and monitoring, including sampling and analysis, are carried out to determine that all applicable requirements are being met. Reports of such inspections should be prepared and circulated as required by the competent authority.

5.8. Transfer of Ownership. The seller of land affected by uranium refining wastes shouldfully inform the buyer and the competent authority of the nature of the wastes. The competent authority should inform the buyer of his obligations for maintenance and control, and the buyer should assume these obligations when the sale is effected.

5.9. Identification. The competent authority may require identification of burial locations through such means as permanent physical markers and the filing of an accurate civil engineering survey with the property records and others.

6. Responsibilities

The responsibilities for waste management may vary considerably from country to country, but it is believed that a basic approach should be capable of being implemented in any situation.

6.1. Responsibilities of the manager and owner/operator.

The manager and owner/operator should, at all times, be subject to the direction of the competent authority, and in particular be directly responsible for :

- (a) Preparation, for approval by the competent authority, of an assessment of potential environmental impacts of the wastes generated and released by the proposed uranium refining operation, including both the initial operation, any subsequent changes, and decommissioning.
- (b) Obtaining all necessary authorizations from the competent authority including those for disposing of wastes and the environmental monitoring programme;
- (c) Establishing and implementing approved effluent release controls and environmental monitoring programmes including pre- and post-operational surveys and keeping all necessary records;
- (d) Ensuring that all waste disposal operations are carried out in accordance with such authorizations and approvals;
- (e) Assessing the consequences of unplanned events which may release additional wastes to the environment, developing approved emergency plans to deal promptly with such events, implementing the plan if

such an event occurs and promptly reporting the event and the action taken to the competent authority;

- (f) Stabilizing the waste retention systems by approved methods during both the operational phase and after operations cease, and taking such other actions as may be deemed necessary by the competent authority;
- (g) Planning the decommissioning of the refinery site, obtaining all necessary authorizations from the competent authority and performing the work in accordance with such authorization;
and
- (h) Carrying out such inspections and keeping such records as may be required by the competent authority.

6.2. Responsibilities of the Competent Authority.

The competent authority is directly responsible for :

- (a) Regulating and controlling the activities of, and giving appropriate direction to, managers and owners/operat
- (b) The prompt review of all proposals submitted by the manager/owner/operator and the issuing of discharge and other authorizations to ensure the protection of the public and the environment. In particular, the level of protection of individuals and the population against the effects of ionizing radiation should comply with the current recommendations of the International Commission on Radiological Protection (ICRP);
- (c) Regulating and inspecting the waste management operations and records to ensure compliance with the authorizations, and

(d) Assessing the consequences of any unplanned event which may cause the release of waste in excess of the authorization and advising the manager/owner/operator of any remedial action which may be necessary to limit the effect of the release and/or prevent a recurrence.

6.3. The division of responsibility between the manager and the owner/operator has not been strictly identified as it is considered that this may vary from one situation to another, according to the type of ownership, amount of government participation, etc. The competent authority should evaluate this situation and clearly establish where the responsibilities lie. In general, it appears to be common practice that the manager is responsible during the operational phase, and that the owner assumes responsibility after the site is decommissioned.

Where the 'operator' is not the 'owner', it is essential that the 'owner' be made fully aware of the probable extent of his future obligations so that he can negotiate such agreements with the operator as are necessary to protect his interests.

6.4. Guidance on the preparation of environmental impact assessments, if required, may be available from the competent authority. The objectives and design of environmental monitoring programmes for radioactive contaminants are given in Chapter 4.

6.5. It is essential to the achievement of their common goal, the safe and economical management of waste, that the relationship between the competent authority and the operating organization should be based on mutual understanding and respect.

References:

- [1] American National Standards Institute, "Stabilization of Uranium-Thorium Milling Waste-Retention Systems", ANSI N313 - 1974 (1974)
- [2] Chemical Week, "Uranium Waste as Fertilizer?", McGraw-Hill, pp. 43-44, (Oct. 24, 1979)
- [3] Clark, F. E., Francke, H. C., and Stohecker, J. W., "Biological Denitrification of High Nitrate Waste Solutions", Proc. 30th Industrial Waste Conference (1975).
- [4] Alexander M., "Denitrification", Introduction to Soil Microbiology, John Wiley & Sons, N.Y. pp 293-308 (1951)
- [5] Prakasam T. B. S., Joo D., Srinath E. G., and Loehr R. C., 29th Annual Purdue Industrial Waste Conference, Indiana (1974).
- [6] Christianson C. W., Rex E. H., Webster W. M., and Virgil F. A., "Reduction of Nitrate Nitrogen by Modified Activated Sludge" USAEC TID-7517. (1967)
- [7] McCarty P. L. and St. Amont P., "Biological Denitrification of Waste Waters by Addition of Organic Materials" Proc. 24th Industrial Waste Conference (1969).
- [8] Barth E. F., Brenner R. C., and Lewis R. F., "Chemical - Biological Control of Nitrogen and Phosphorus in Wastewater Effluent", Water Pollution Control Federation, V-40 (12) pp. 2040-2054 (1968).
- [9] NLCO-1110, "Study for Disposal of Uranium Contaminated Ferrous Scrap, Shaw W. E. (April 1974)
- [10] NLCO-1113, "Uranium DEcontamination of Common Metals by Smelting", Mautz E. W., et al (Feb. 1975)

- [11] NLCO-1128, "The Ferrous and Common Nonferrous Metals Industries and Associated Scrap Metals", Mautz E. W., Editor (Nov. 1975)
- [12] NLCO-1132, "Feasibility Study of a Portable Smelter for Scrap Metals", Cavendish J. H., Editor (June 1976)
- [13] ORNL/TM-6442, "Generic Assessment of Radiation Exposures to Workers in a Portable Smelter and to the Surrounding Population", Randolph M. L., et al (Oct. 1978)

Bibliography

Harrington, C. D., Ruehle A. E., "Uranium Production Technology", D. Van Nostrand Co. Inc., Princeton, N. J. (1959)

American Industrial Hygiene Association, "Industrial Ventilation - A Manual of Recommended Practice" 16th Edition, AIHA, Lansing, Michigan (1980)

International Commission on Radiation Protection, "Recommendations of the International Commission on Radiation Protection" ICRP Publication 26 (Adopted January, 1977)

National Council on Radiation Protection and Measurements, "Basic Radiation Protection Criteria", NCRP Report No. 39 (January 1971)

U. S. Atomic Energy Commission, "Environmental Survey of the Nuclear Fuel Cycle", Section C-1, USAEC, Fuels and Materials and Directorate of Licensing, Washington DC (1972)

National Research Council, "The Shallow Land Burial of Low-Level Radioactively Contaminated Solid Wastes", National Academy of Sciences, Washington, D.C. (1976)

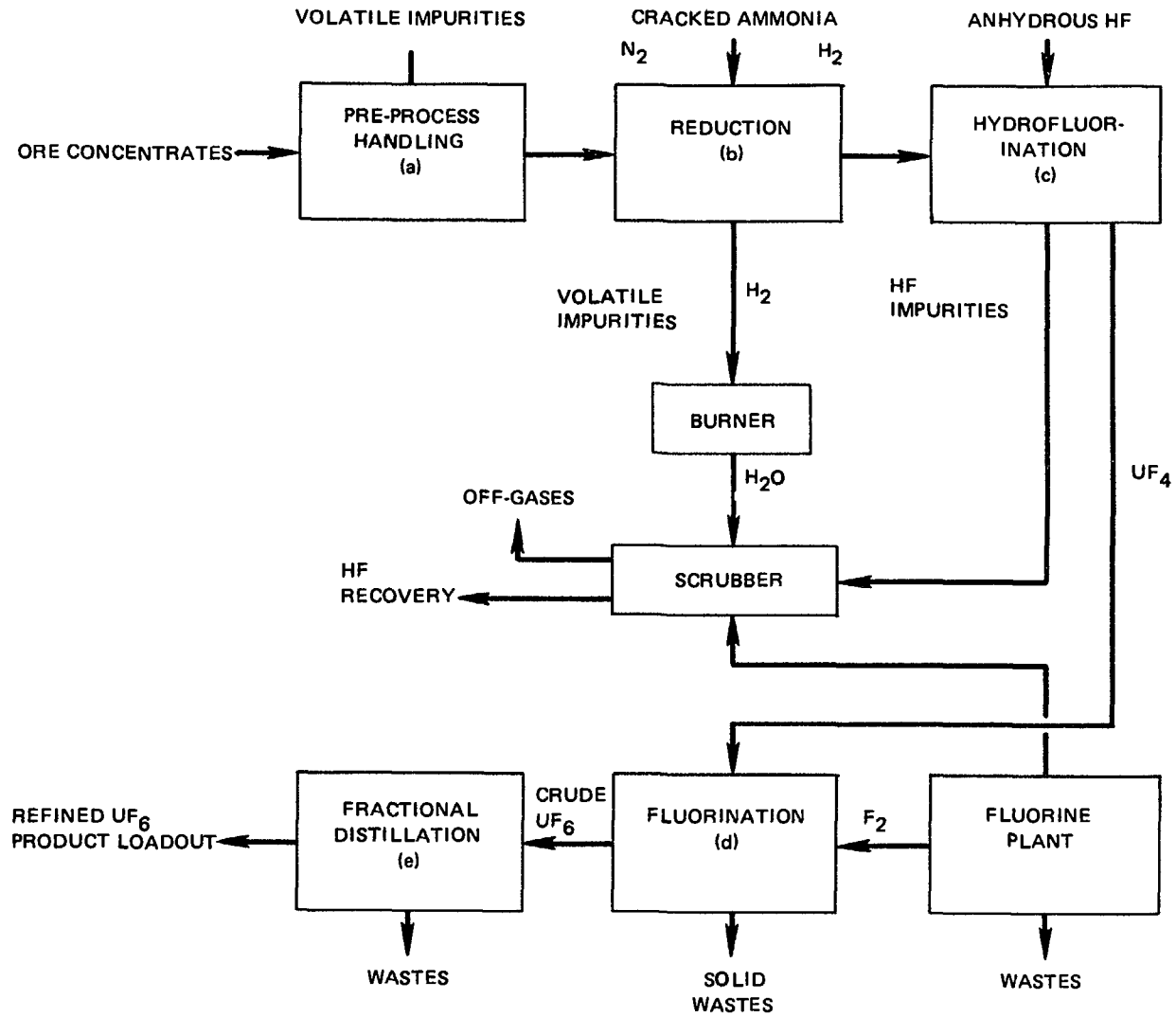
APPENDIX I

FLOW SHEETS

FLOW SHEETS

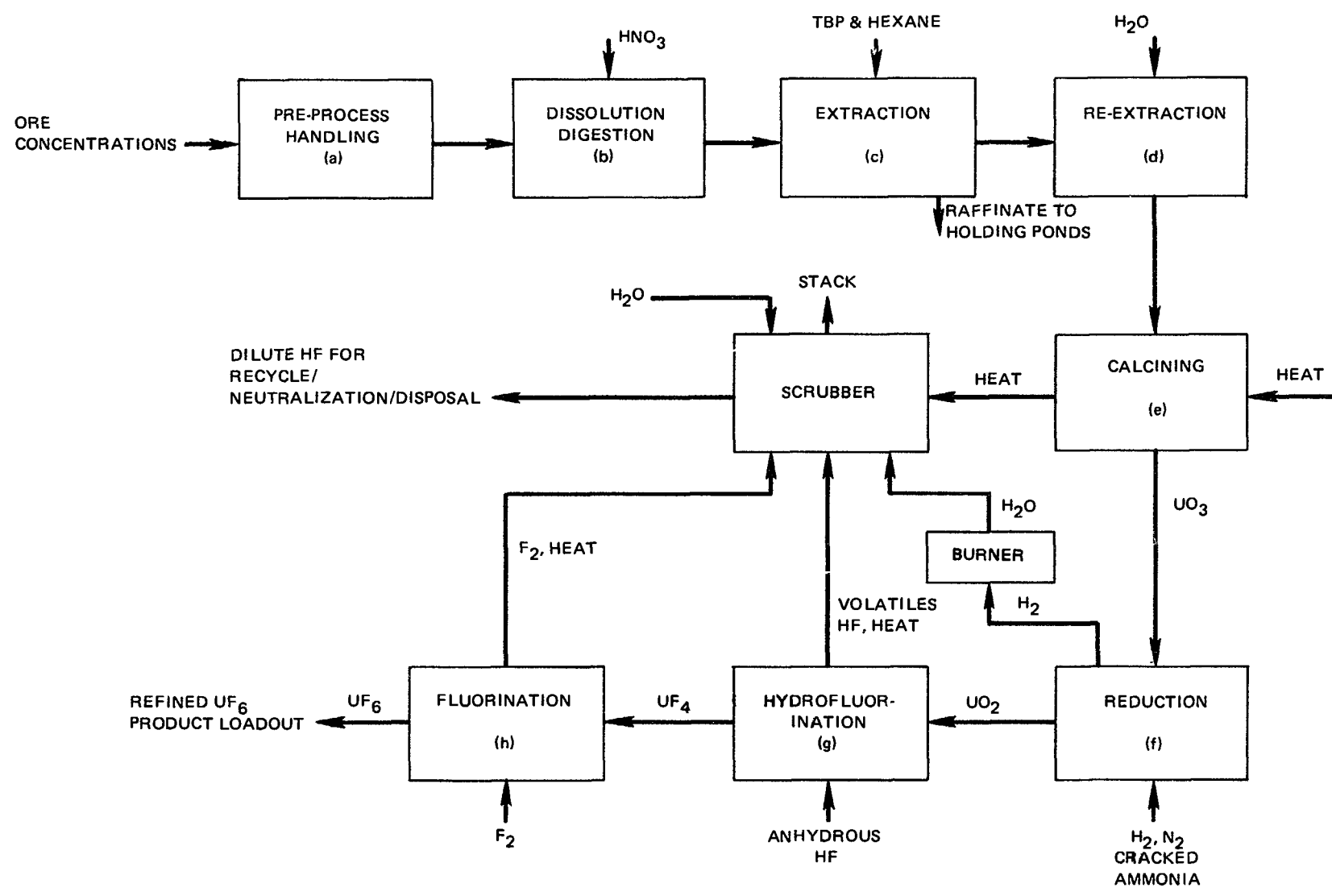
1. TYPICAL DRY HYDROFLUOR PROCESS	PAGE	39
2. TYPICAL WET SOLVENT EXTRACTION— FLUORINATION PROCESS	PAGE	40
3. ALLIED CHEMICAL	PAGE	41
4. COMURHEX	PAGE	42—47
5. ELDORADO NUCLEAR LIMITED	PAGE	48—61
6. KERR-McGEE NUCLEAR CORPORATION	PAGE	62

1. Flow Sheet Listing (1. through 6.)



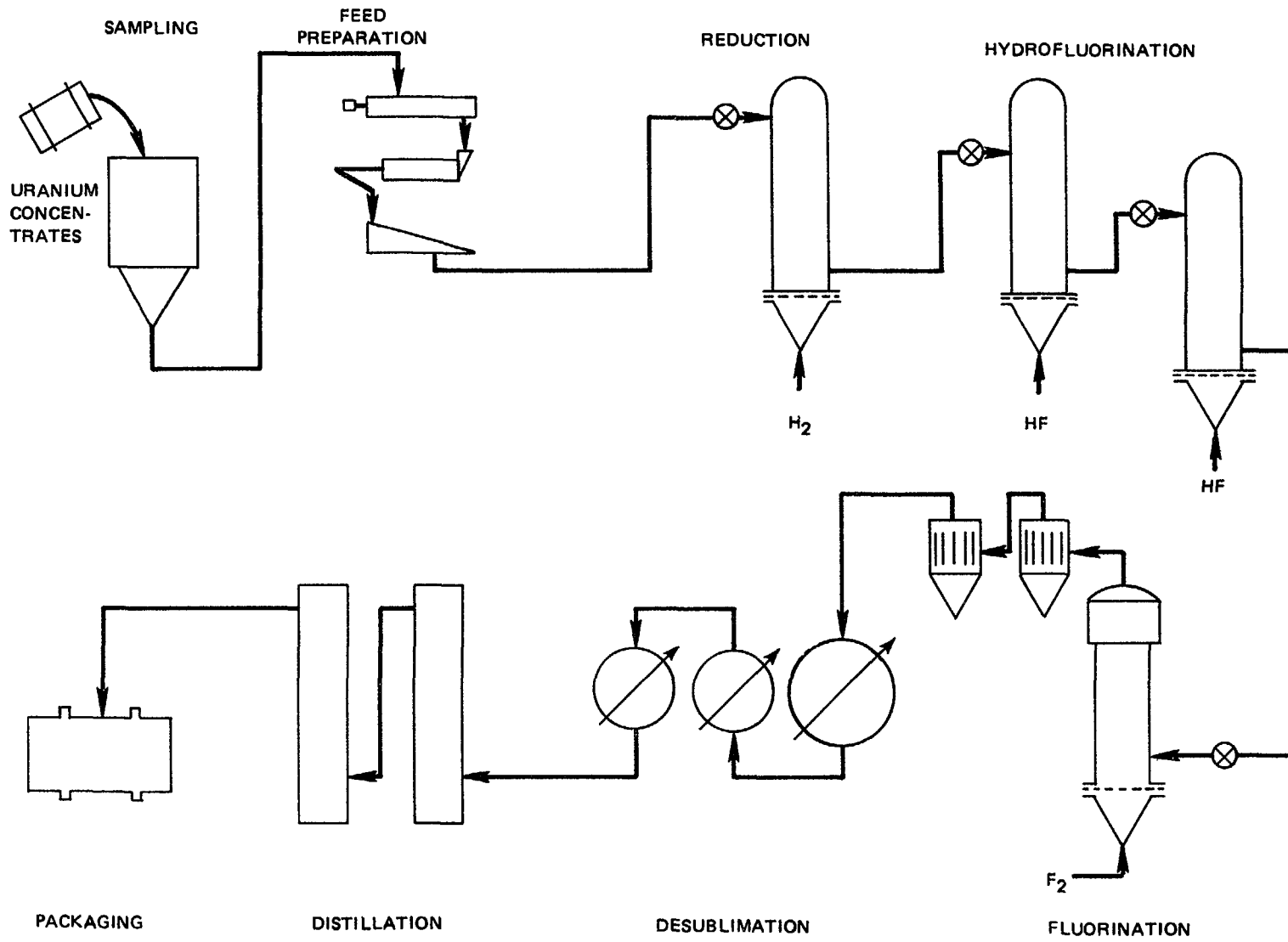
UF₆ PRODUCTION - HYDROFLUOR PROCESS
BLOCK DIAGRAM

3. UF₆ Production - Wet Solvent Extraction - Fluorination -
Block Diagram

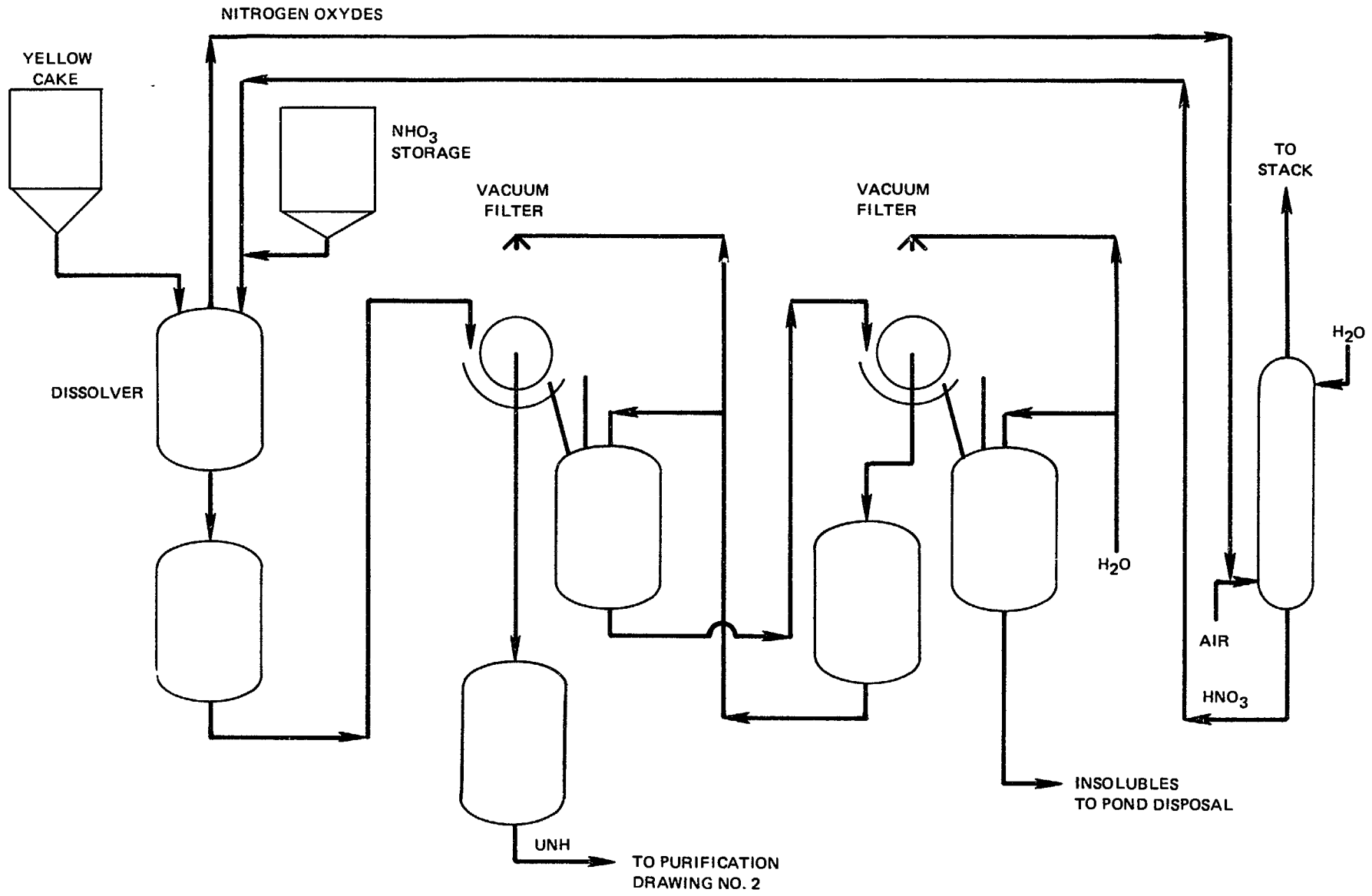


UF₆ PRODUCTION - WET SOLVENT EXTRACTION - FLUORINATION
BLOCK DIAGRAM

FLUORIDE VOLATILITY PROCESS



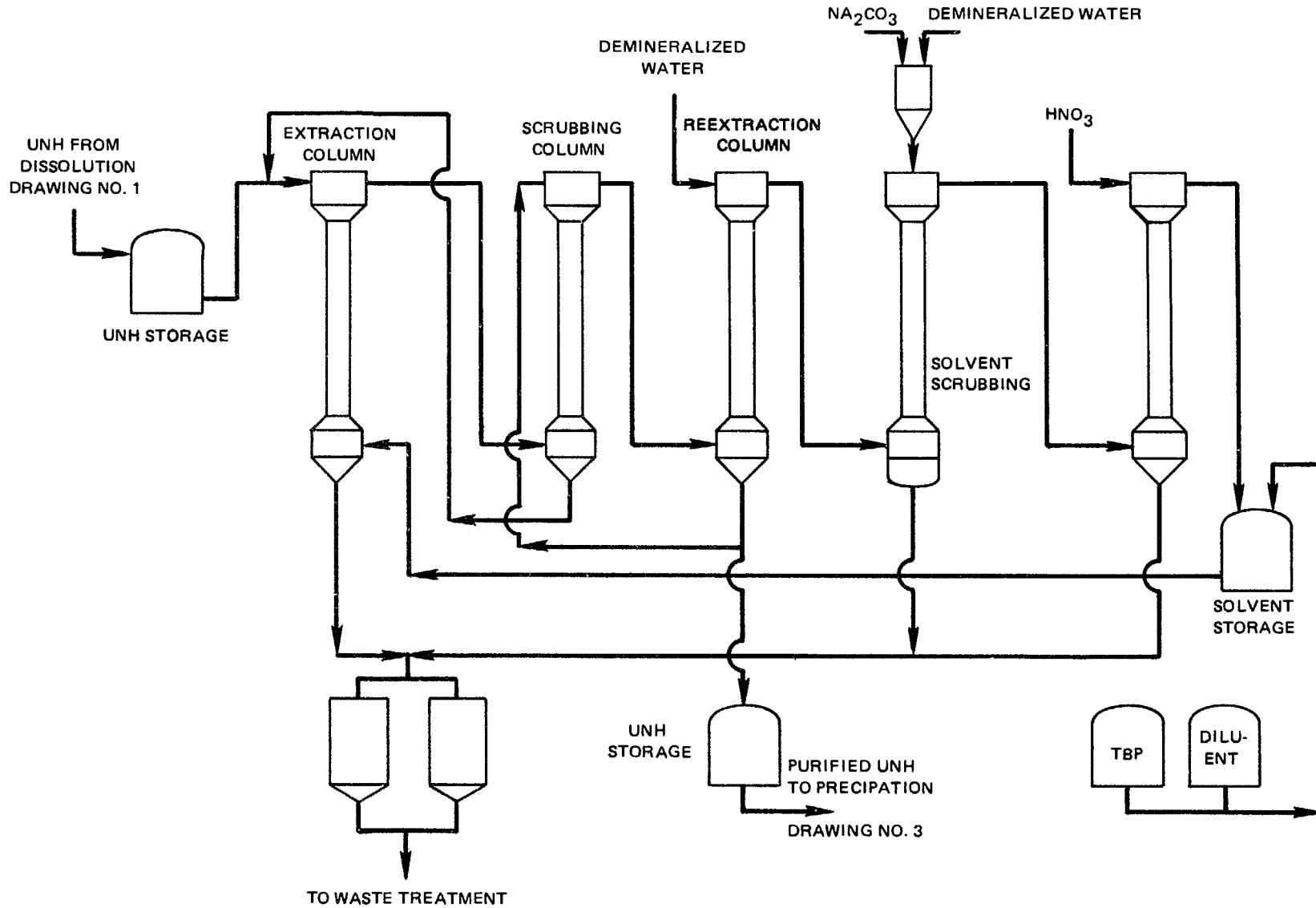
ALLIED CHEMICAL



5. COMURHEX Drawing No. 1

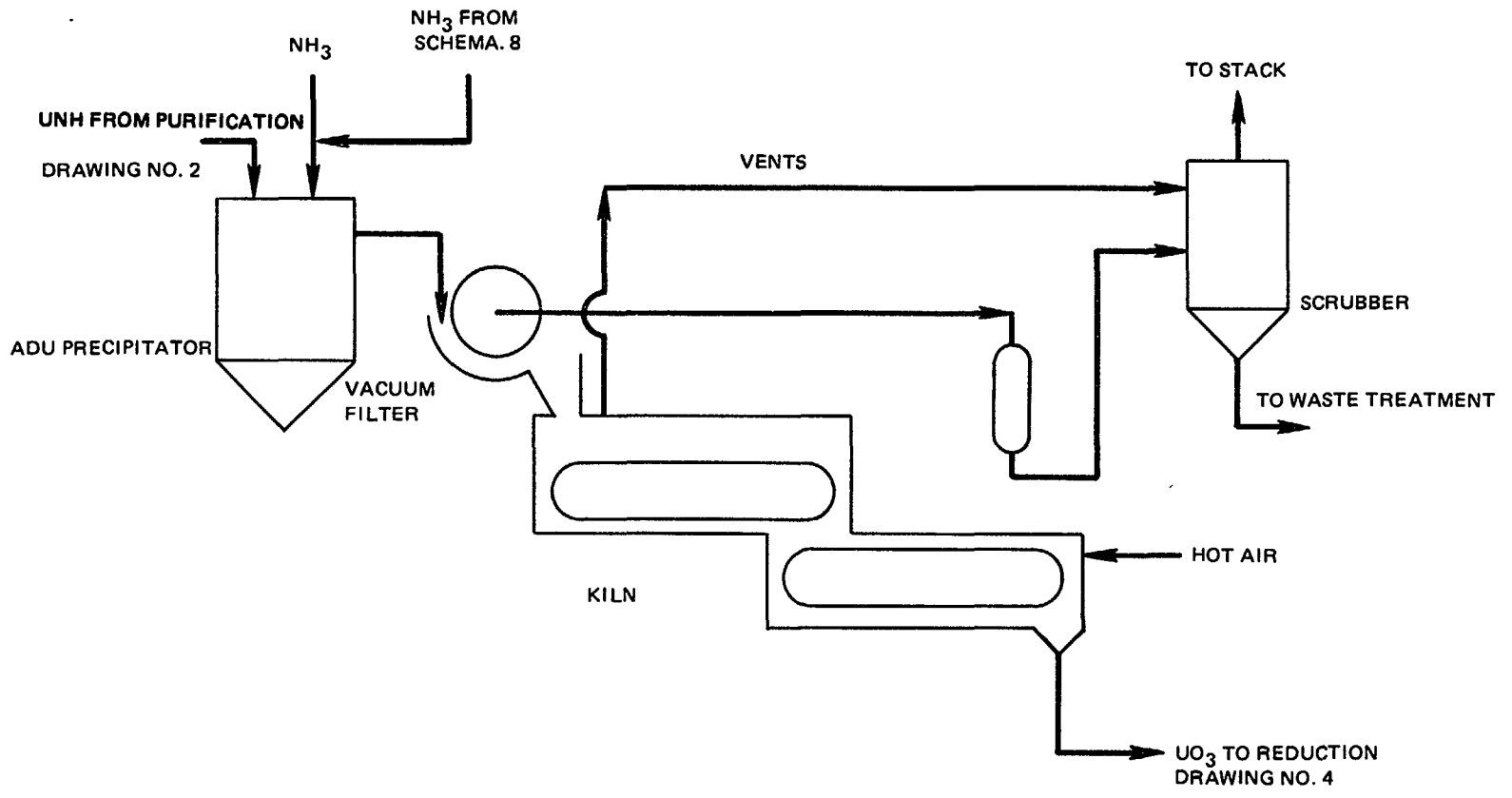
42

COMURHEX DRAWING NO. 1

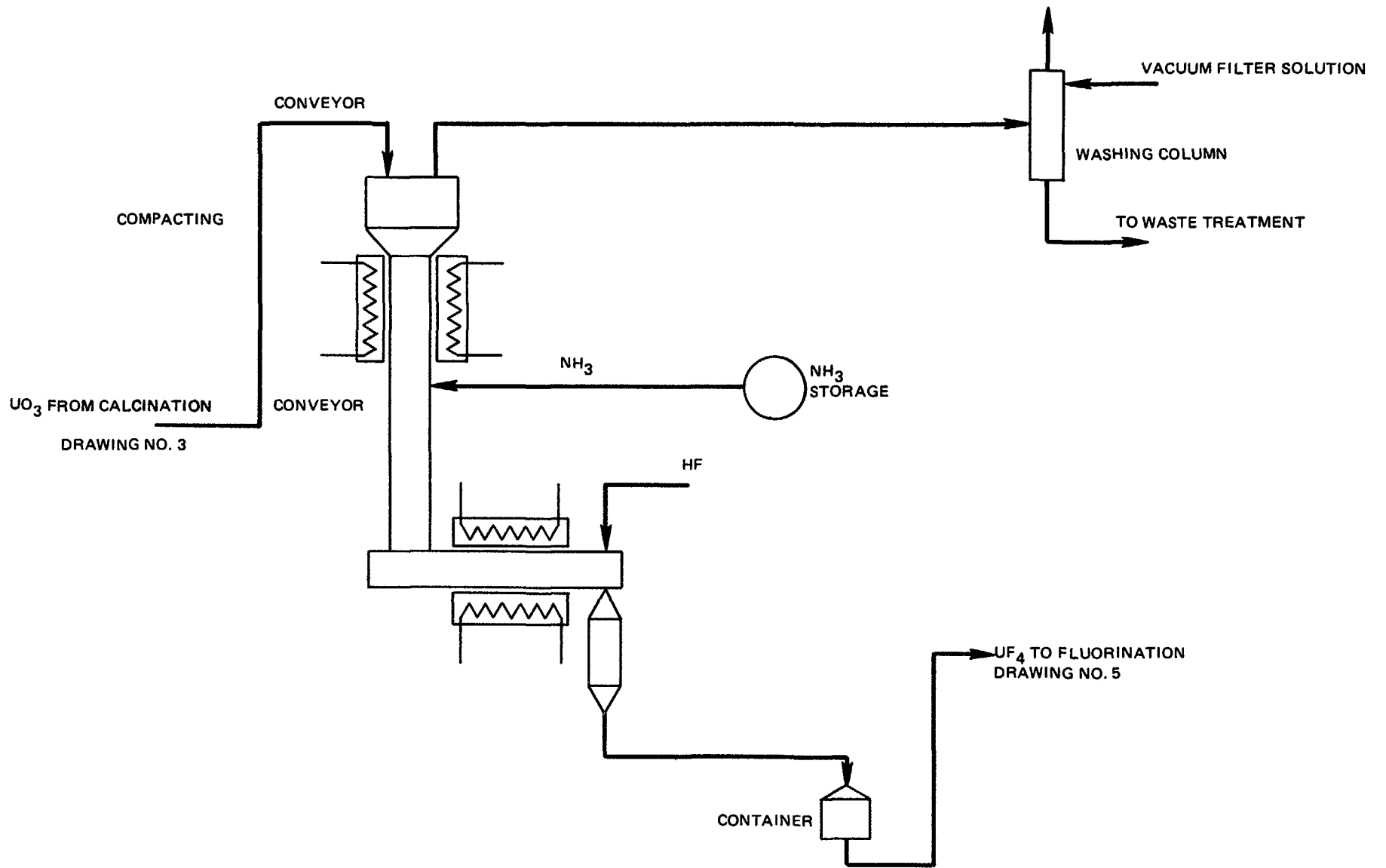


COMURHEX DRAWING NO. 2

6. COMURHEX Drawing No. 2

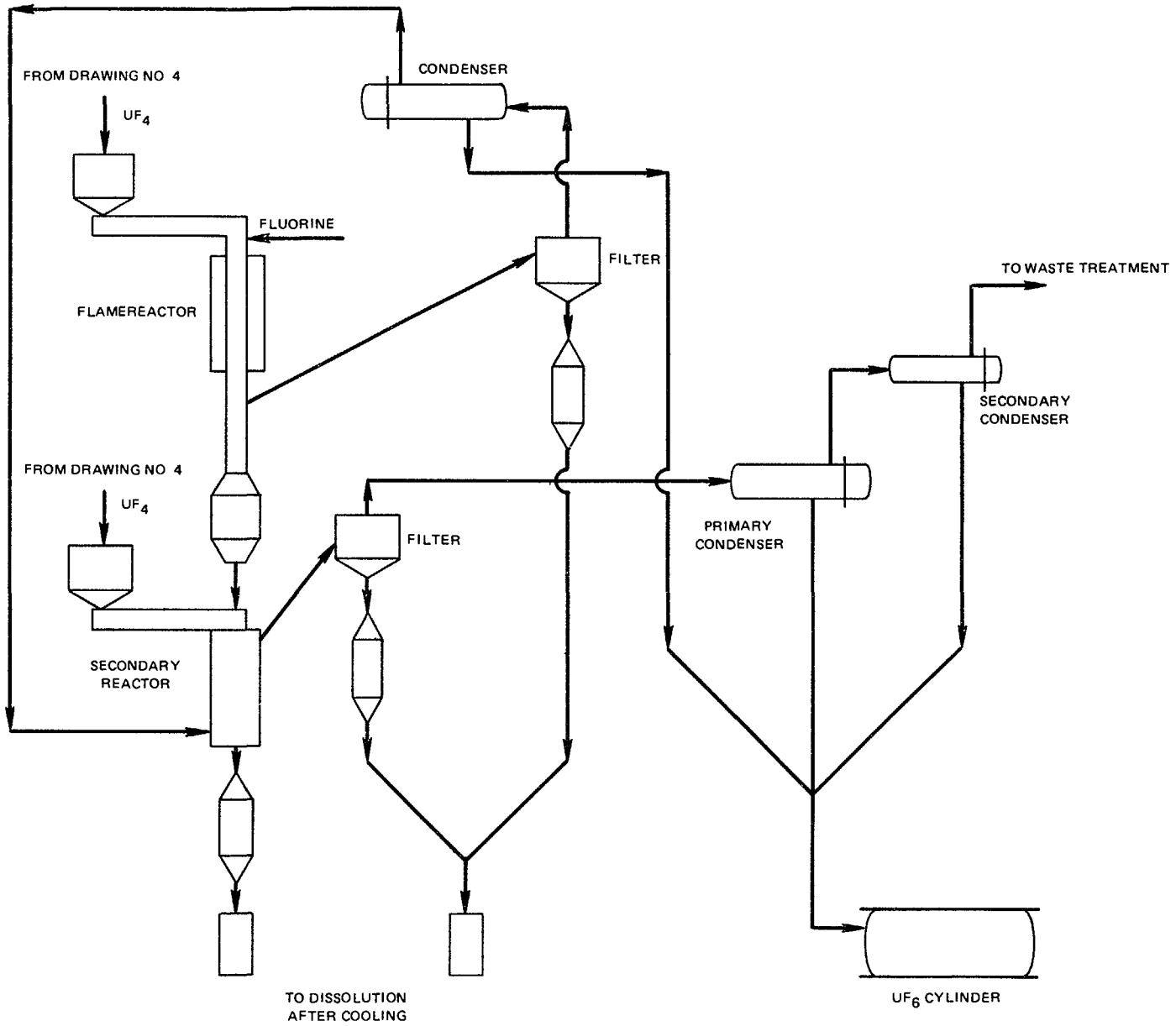


COMURHEX DRAWING NO. 3

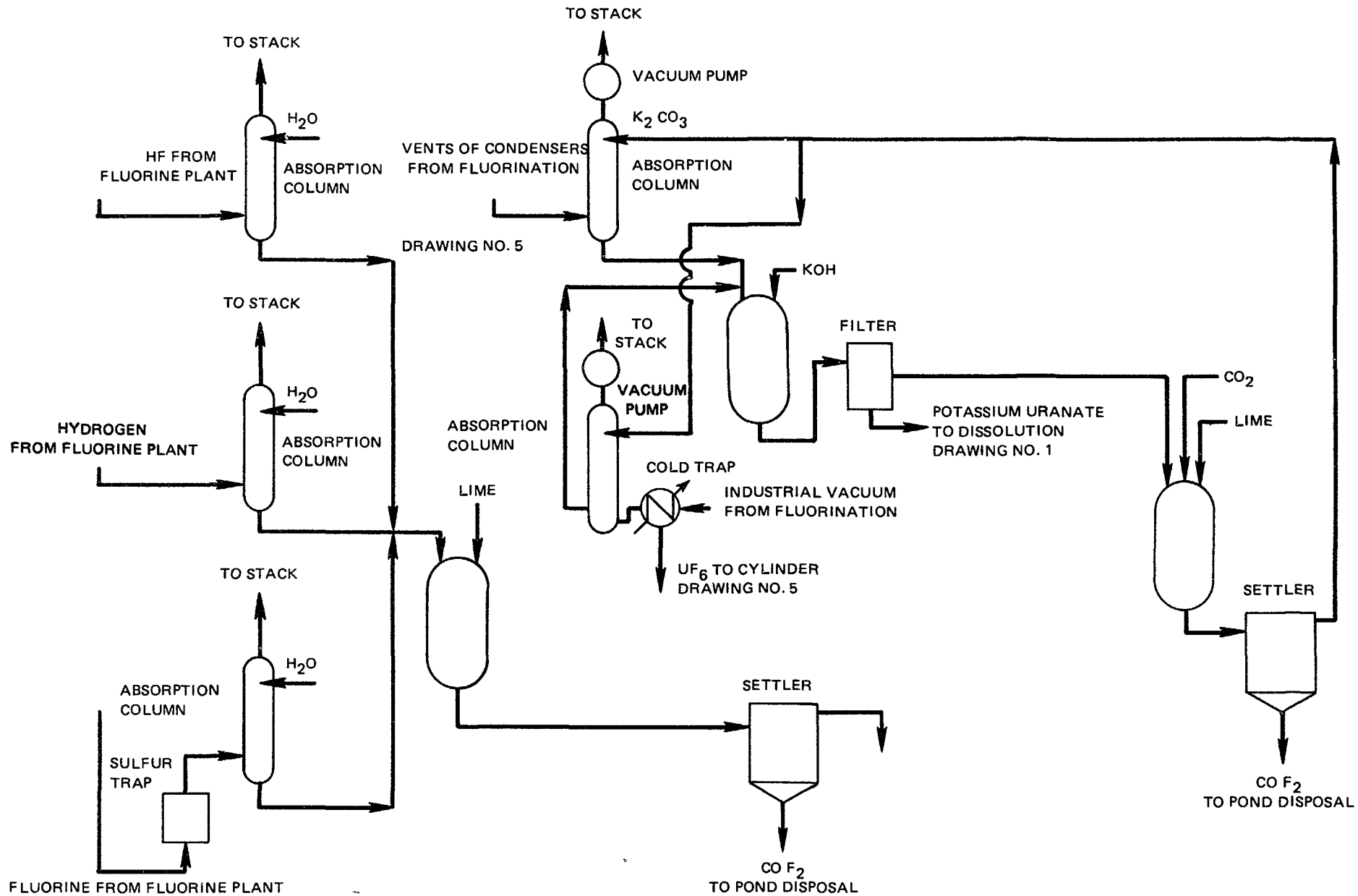


COMURHEX DRAWING NO. 4

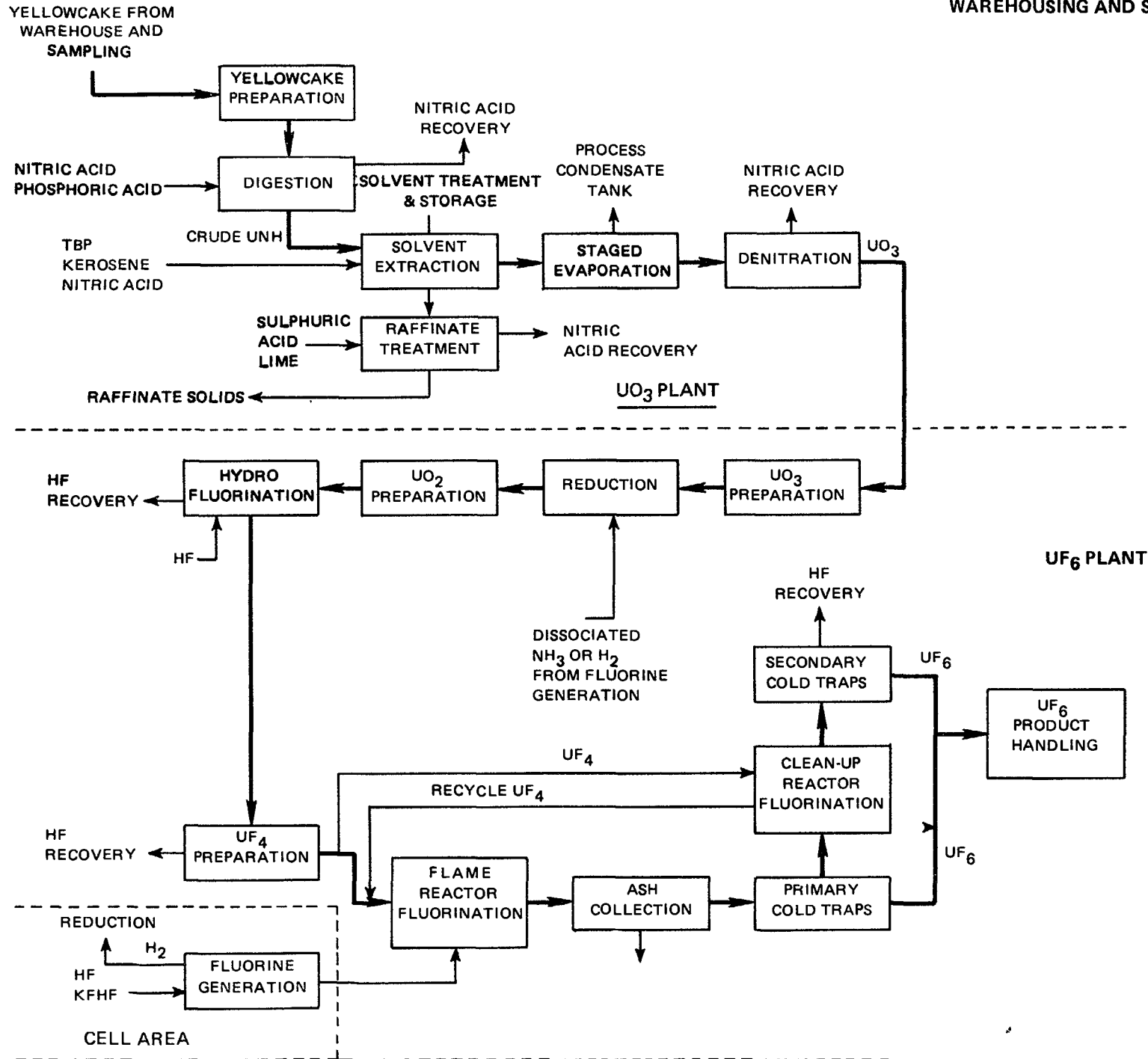
8. COMURHEX Drawing No. 4



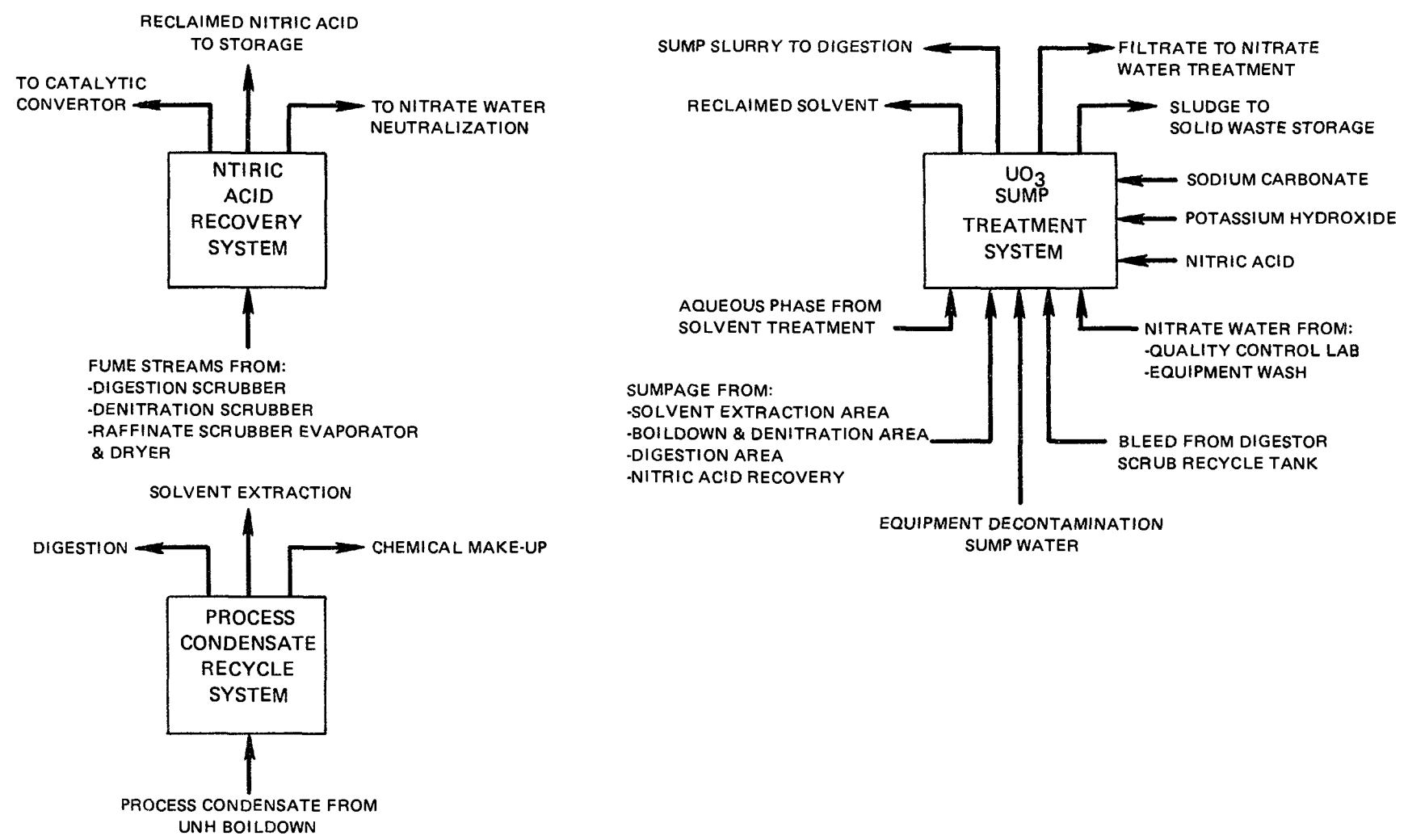
COMURHEX DRAWING NO. 5



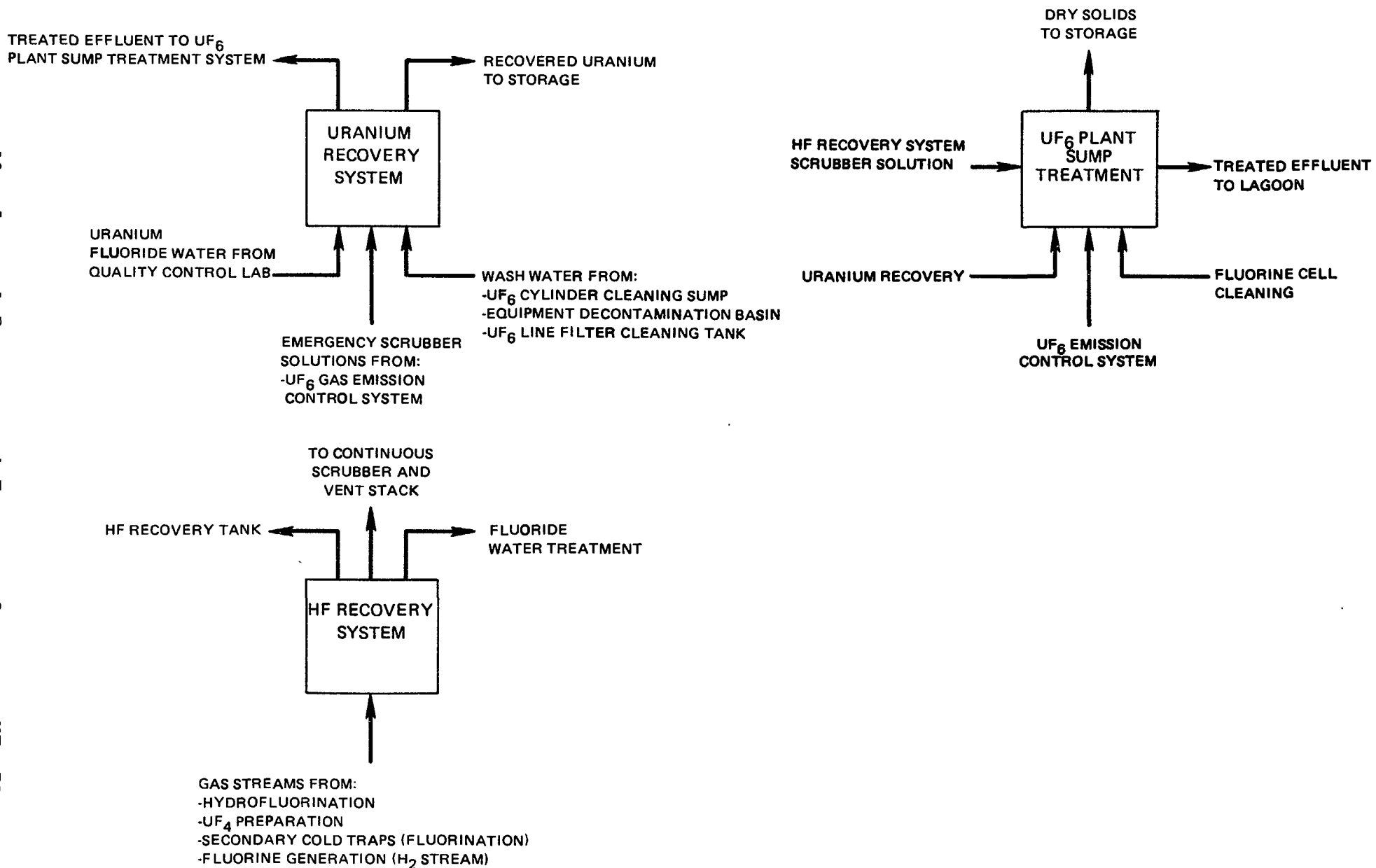
COMURHEX DRAWING NO. 6



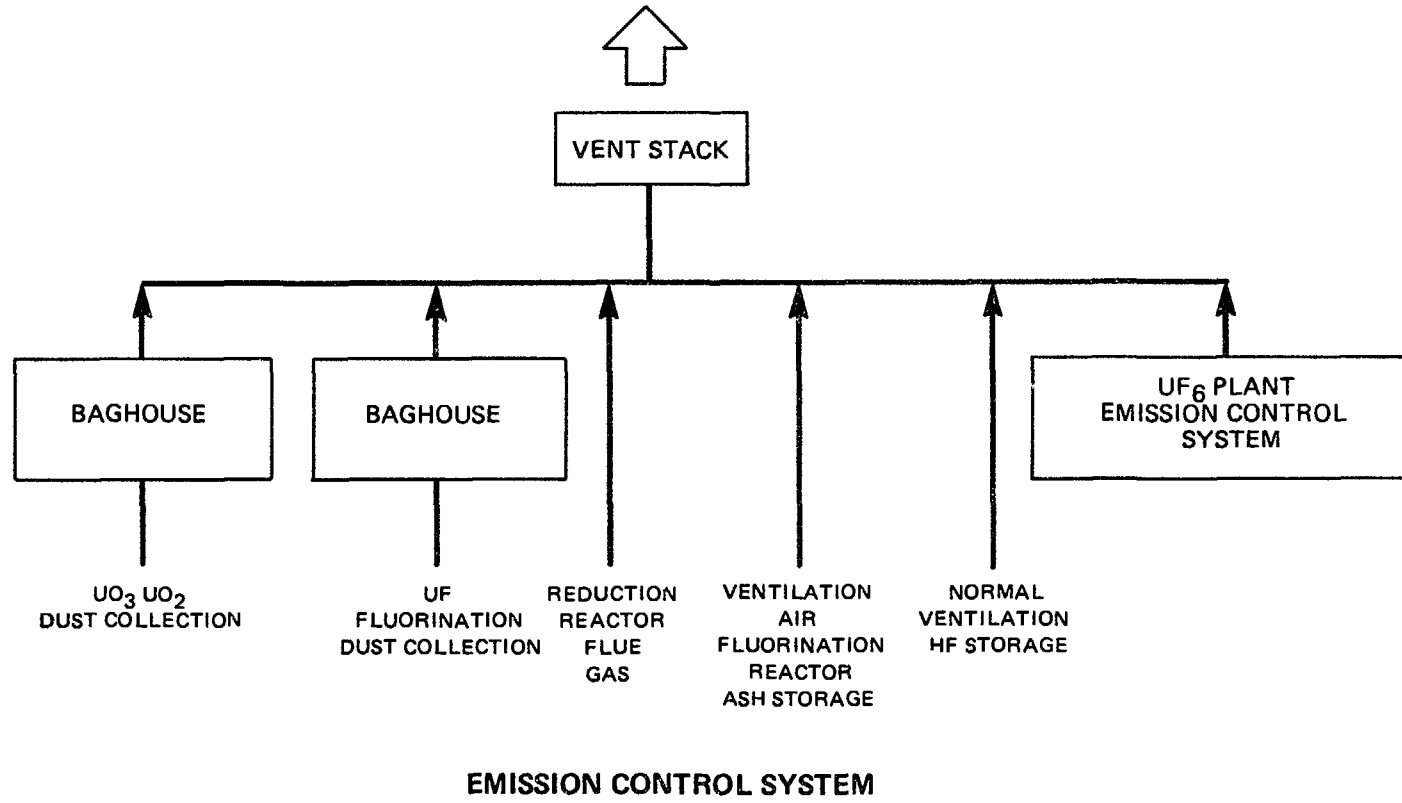
11. Warehousing and Sampling/UF₆ Plant



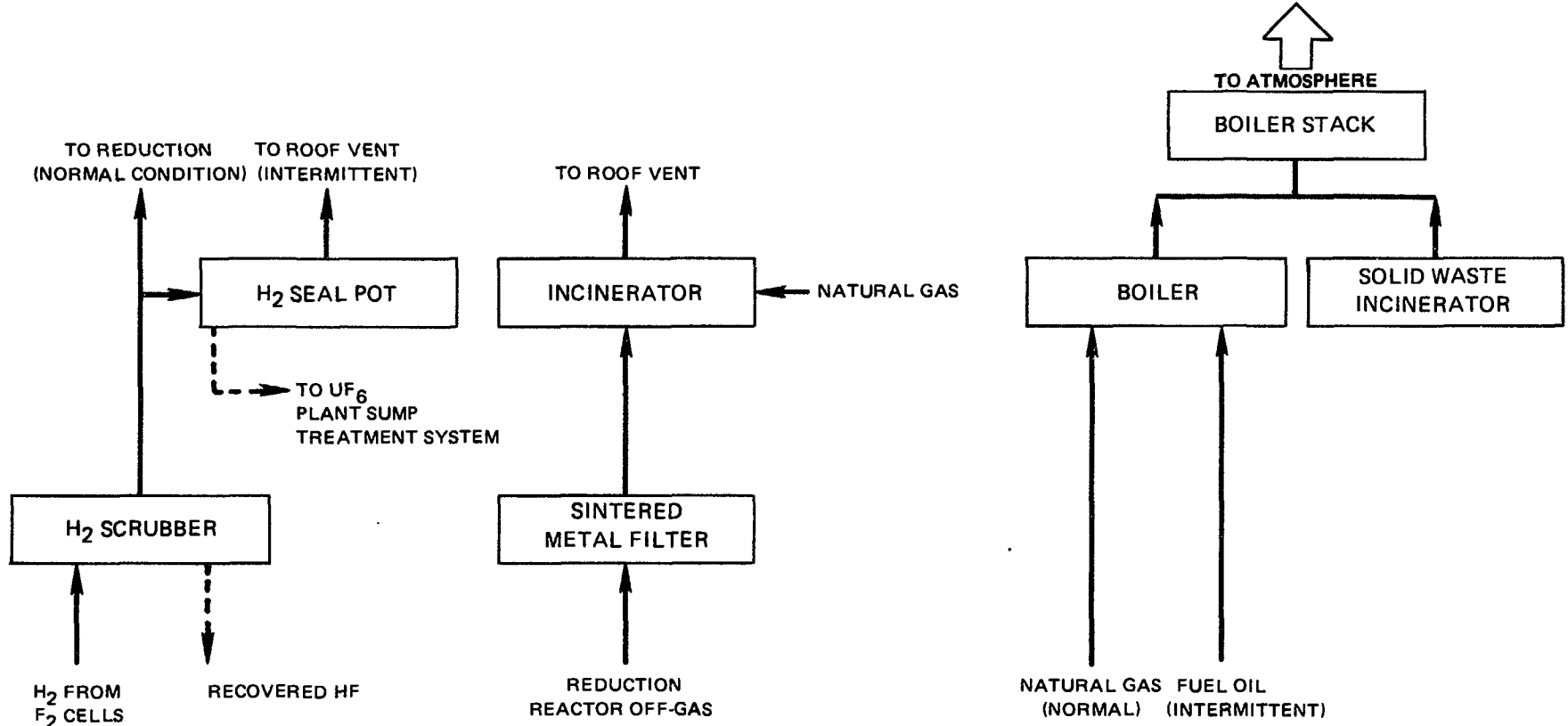
INTERNAL RECOVERY AND TREATMENT SYSTEM-UO₃ PLANT



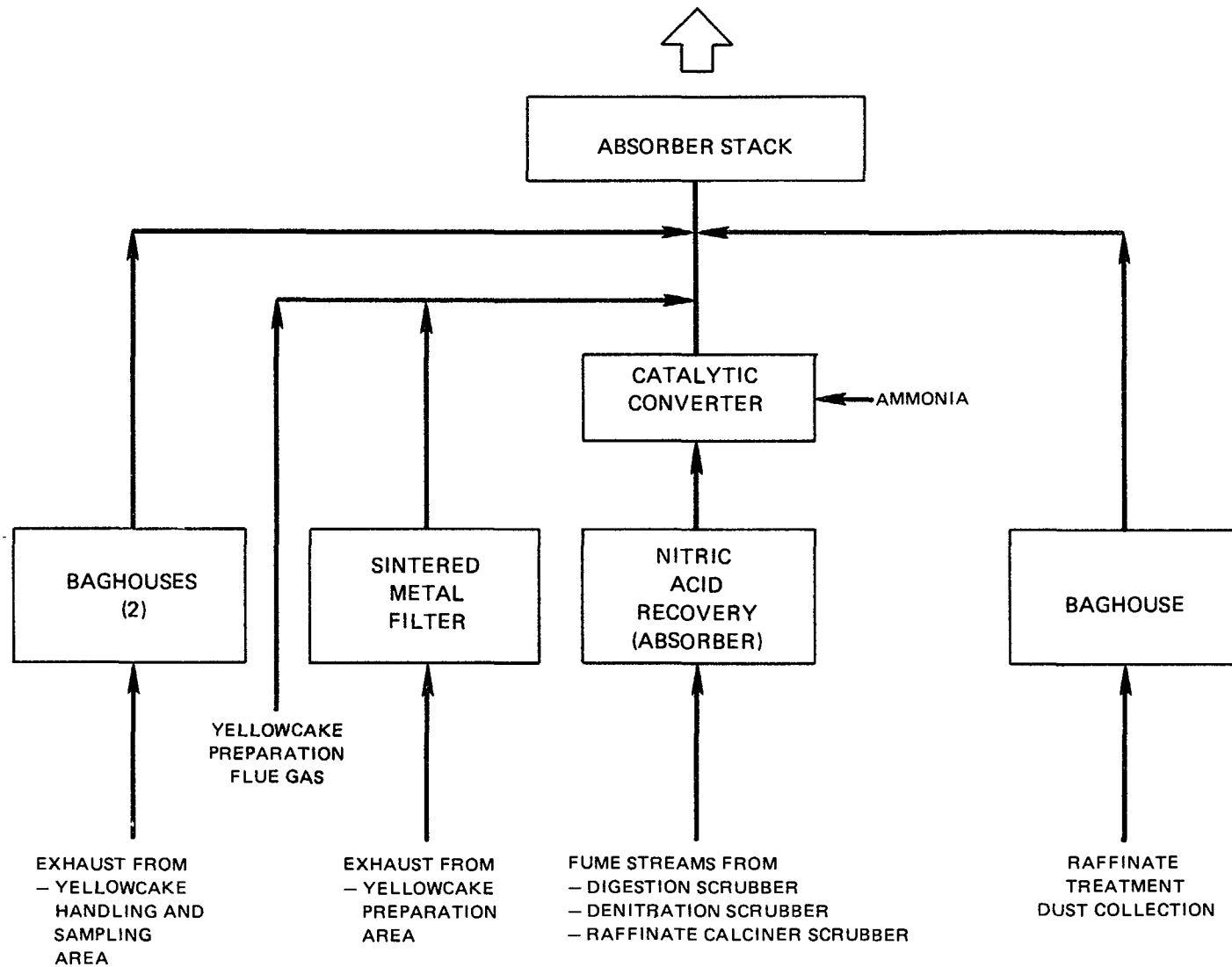
INTERNAL RECOVERY AND TREATMENT SYSTEMS UF₆ PLANT



15. Emission Control System - Boiler Stack



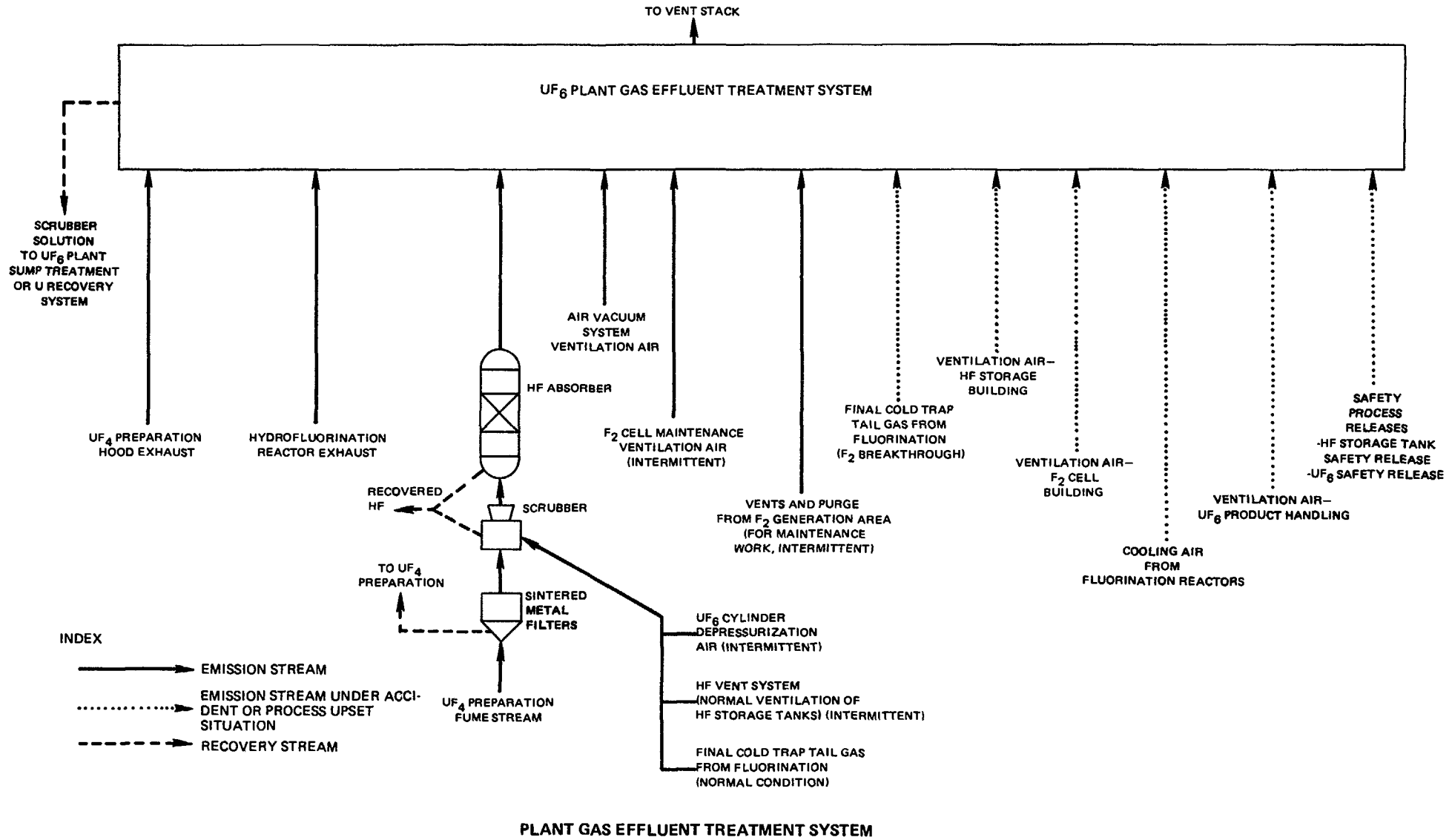
EMISSION CONTROL SYSTEM



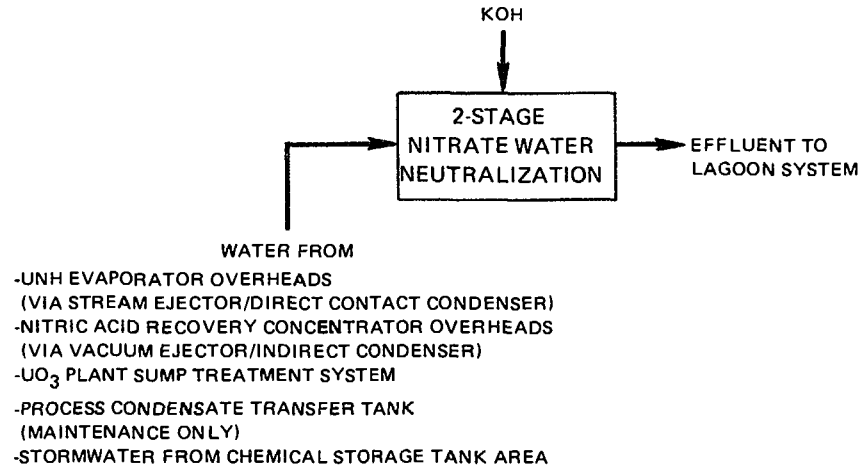
EMISSION CONTROL SYSTEM

17. Plant Gas Effluent Treatment System

S4

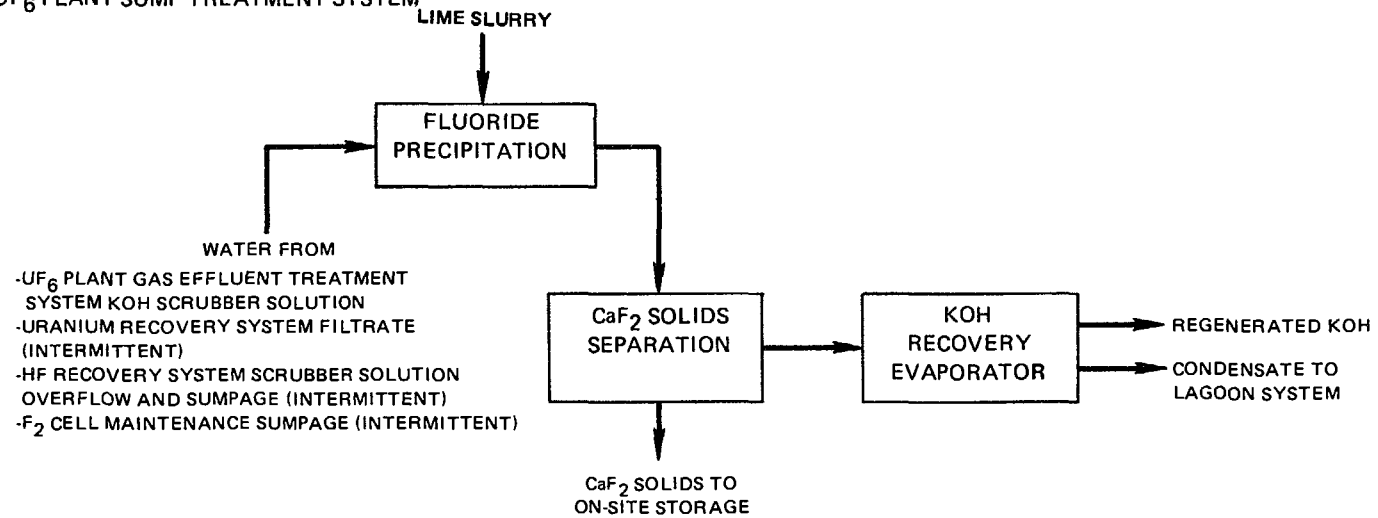


NITRATE WATER TREATMENT



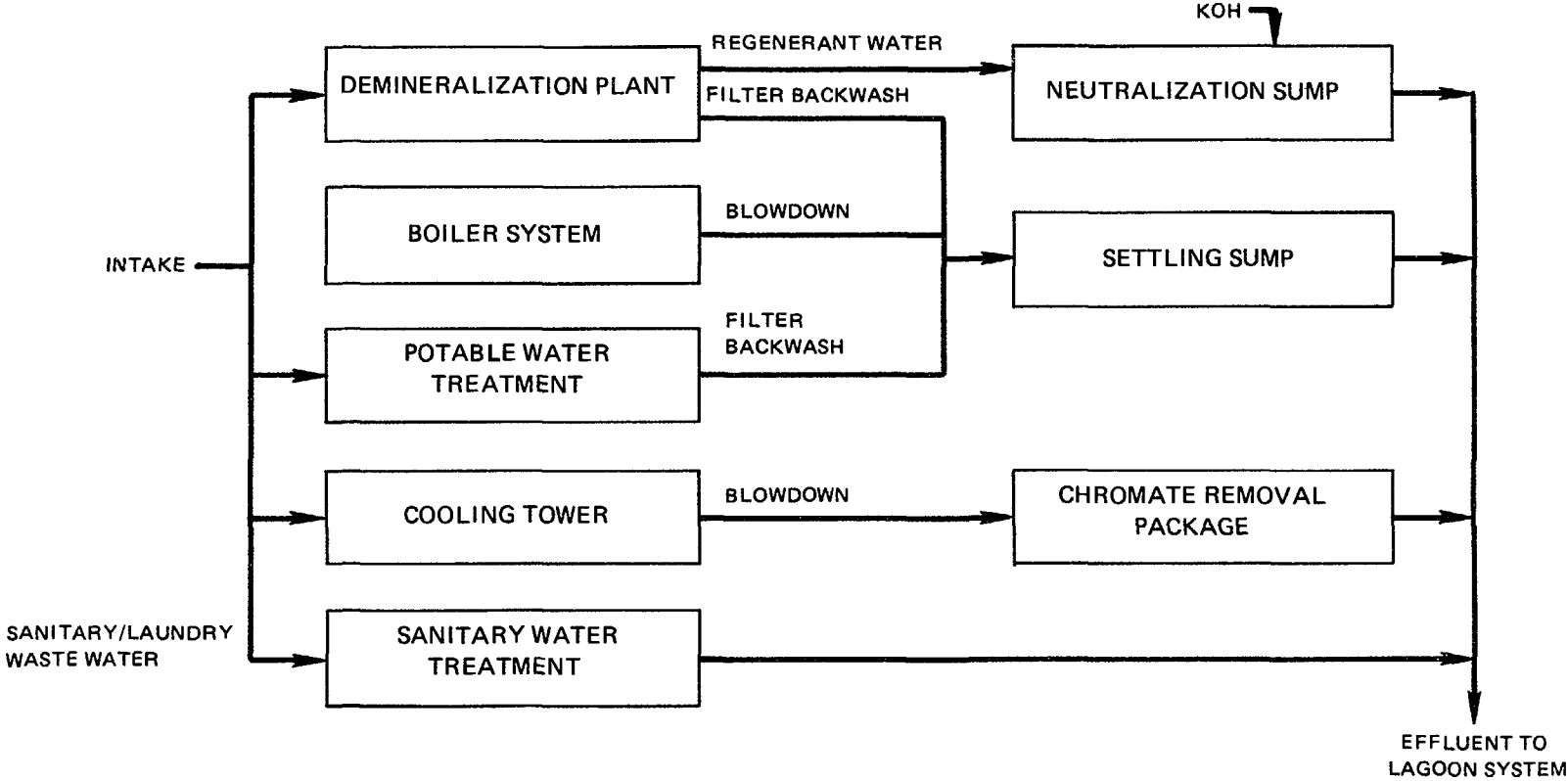
PROCESS EFFLUENT SOURCES

UF₆ PLANT SUMP TREATMENT SYSTEM

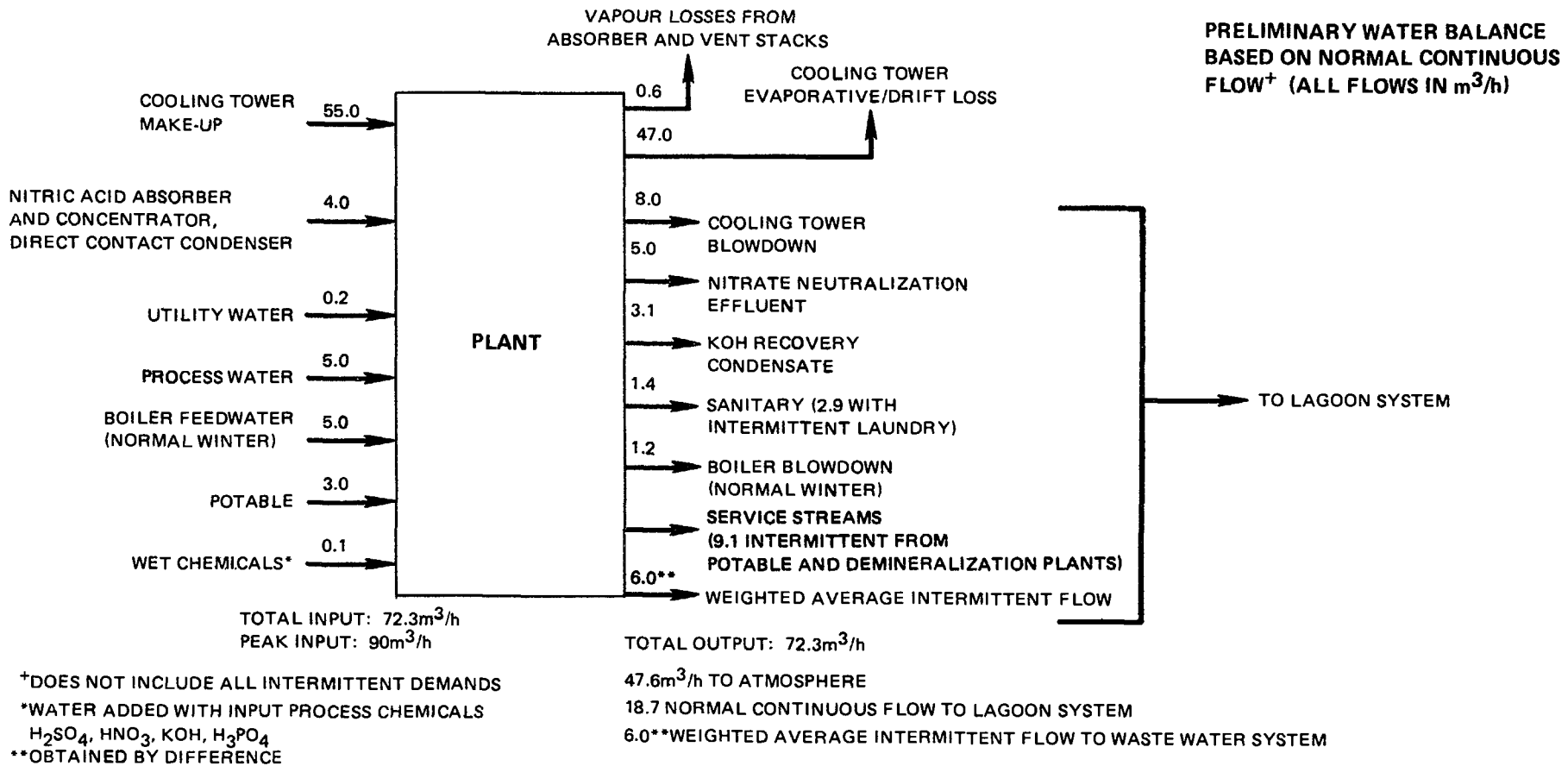


WATER MANAGEMENT SCHEMATIC

**SERVICE AREA
EFFLUENT SOURCES**

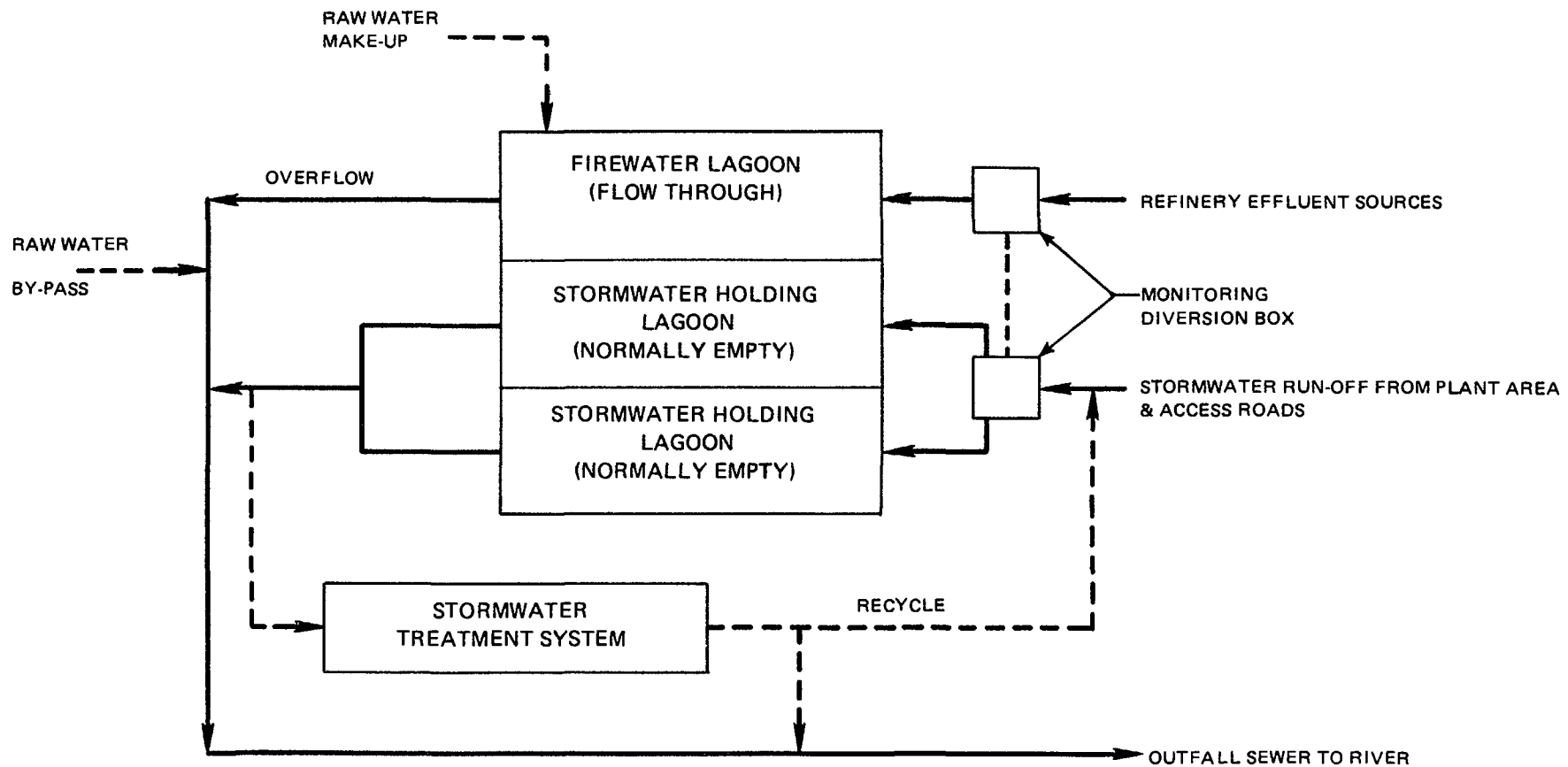


WATER MANAGEMENT SCHEMATIC



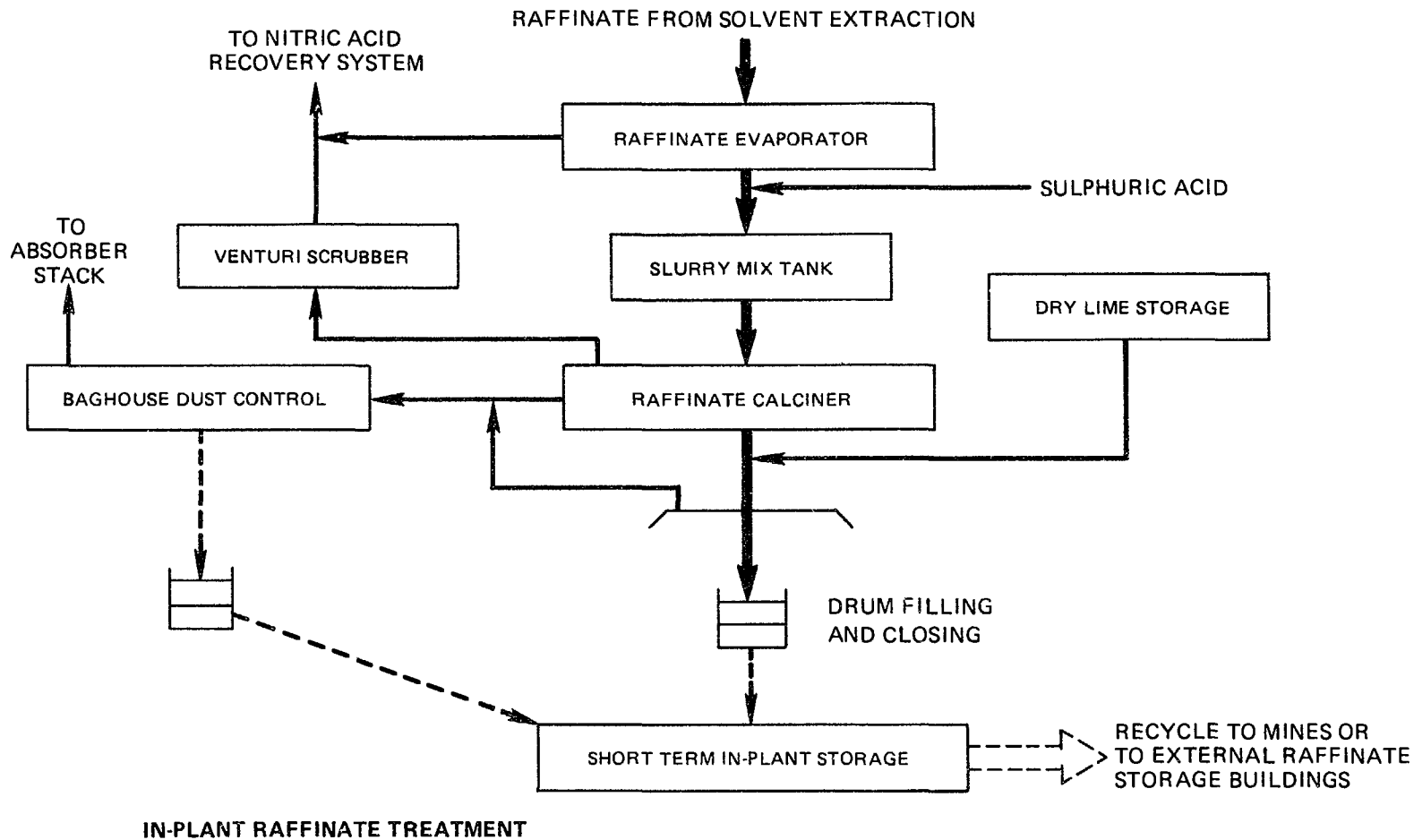
WATER MANAGEMENT SCHEMATIC

LAGOON SYSTEM



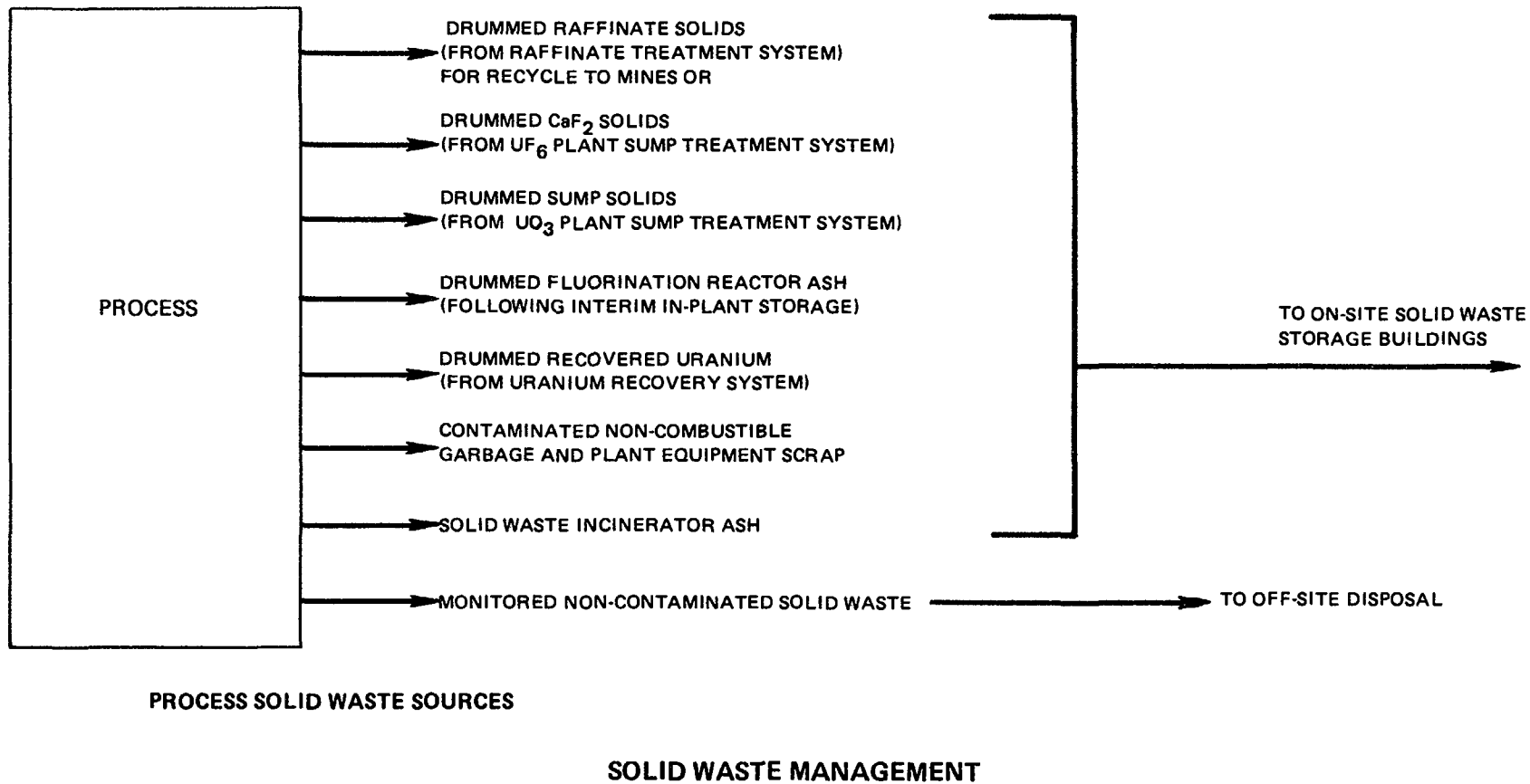
WATER MANAGEMENT SCHEMATIC

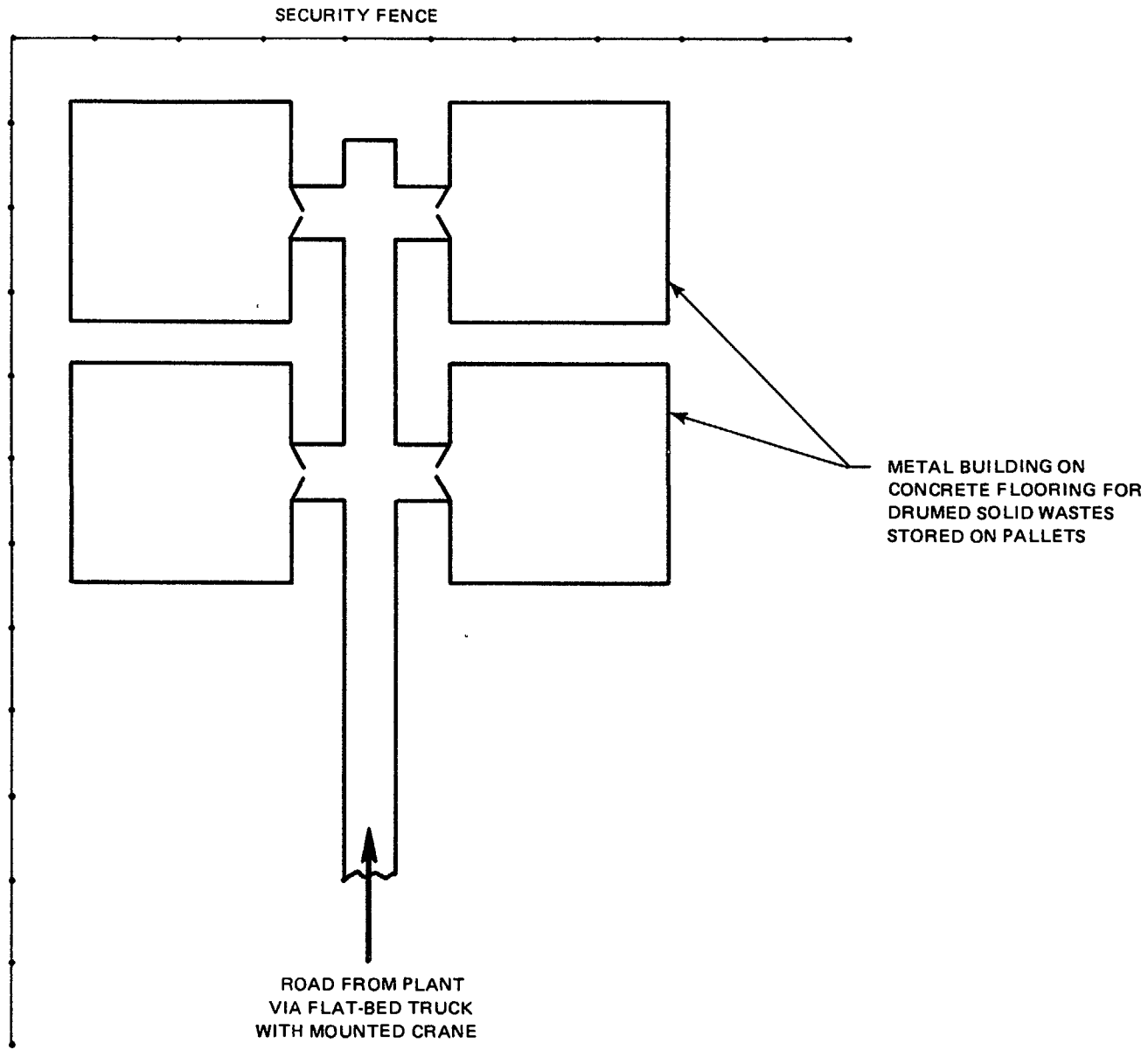
21. Water Management Schematic - Lagoon System



IN-PLANT RAFFINATE TREATMENT

SOLID WASTE MANAGEMENT

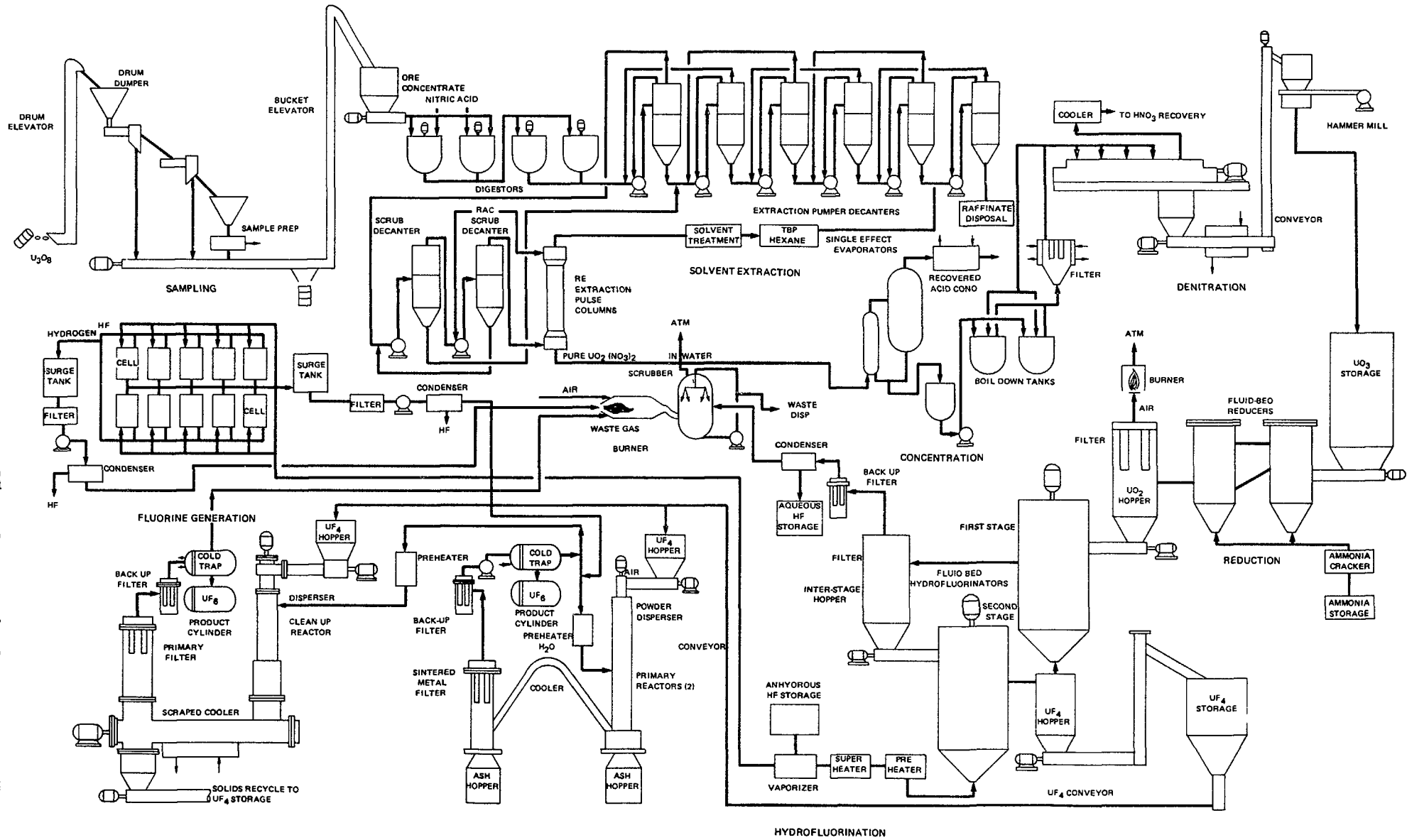




SCHEMATIC OF ON-SITE LOW-LEVEL SOLID WASTE STORAGE COMPOUND

SOLID WASTE MANAGEMENT

SEQUOYAH CONVERSION FACILITY



APPENDIX II

Table of Waste Streams

The data in the following tables represent a range based on approximations of wastes generated by the various processes in use in 1980.

APPENDIX II

Solid Wastes

	<u>Range</u>
U (kg)	to 4000
CaF ₂ (kg)	2 x 10 ⁵ - 12 x 10 ⁶
²³⁸ U daughters (Ci)	0.5 - 6600
²³² Th (kg)	2 - 10 ⁵
²²⁶ Ra (Ci)	4 x 10 ⁻⁶ - 0.5
Heavy metals (kg)	to ~10 ⁴
Scrap metal (kg)	~ 500,000
Incinerator ash (kg)	13,000 - 33,000
Other non-combustible (kg)	~ 300,000
²³⁰ Th (Ci)	0.1 - 50
NO ₃ ⁻ (kg)	0 - 9,000
Silious filter residues (kg)	2-3 x 10 ⁵

Liquid Wastes

	<u>Range</u>
U (kg)	10 - 2500
CaF ₂ (kg)	Small
²³⁸ U daughters (Ci)	0 - 6800
²³² Th (kg)	0 - 20,000
²²⁶ Ra (Ci)	0 - 1
F ⁻ (kg)	0 - 12,000
Heavy metals (kg)	Small
TBP (kg)	0 - 16,000
Diluent (kg)	0 - 54,000
NH ₃ (kg)	0 - 700,000
Na ₂ S (kg)	0 - 9600
²³⁰ Th (Ci)	0 - 100
NO ₃ ⁻ (kg)	0 - 24 × 10 ⁶

Emissions to Atmosphere

	<u>Range</u>
U (kg)	100 - 1200
NO _x (kg)	0 - 160,000
²³⁸ U daughters (Ci)	0.001 - 0.15
²³² Th (kg)	0 - 3.8
²²⁶ Ra (Ci)	to $\sim 5 \times 10^{-5}$
F ⁻ (kg)	100 - 3800
TBP	Trace
Diluent	Trace
NH ₃ (kg)	500 - 10,500
²²² Rn (Ci)	0.5 - 2.0
SO ₂ (kg)	13,000 - 260,000
H ₂ S (kg)	Trace
²³⁰ Th (Ci)	to $\sim 2 \times 10^{-3}$

Definition of Terms Used in the Report

The technical terms used in this Report have their accepted scientific meanings. The definitions of certain terms are as follows:

Activated carbon. Usually a charcoal used to trap airborne contaminants such as organic vapors.

Approved. Approved by the competent authority.

Barren cake. Filter cake from which the recoverable uranium has been removed.

Calciner. Vessel used to produce only a calcined product which can be processed further.

Competent Authority. A national or other authority whose jurisdiction applies to the protection of man and the environment from the activities of the refinery.

Final ash residue. The product of an incineration step after the recoverable uranium has been removed.

Final effluent. Solution which is discharged from the controlled site.

Filter cake. The product from filtering a slurry through a plate and frame or rotary vacuum filter.

Manager. A duly qualified and appointed person responsible for the administration and direction of the refinery.

Non-contact cooling water. Raw or potable water used for cooling process equipment but which has received no chemical additives for corrosion or slime control.

On-site liquid wastes. Within the property boundary of the refinery site.

Operator. Any person, government or other entity that conducts or carries on operations for the refining and conversion of uranium ore concentrate.

Owner. Any person, government or other entity that holds title to or owns the land upon which the plant or other facilities are located.

Spent organic solutions. Typically, these consist of used tri-n-butyl phosphate (TBP), hexane, dodecane, and/or kerosene solutions no longer suitable for process use.

Uranium refining and conversion. All of the steps required to process ore concentrate, usually 60-80% uranium, into uranium hexafluoride (UF_6).

Vessel. Equipment where processing, evaporating, melting or other operations take place.

Waste retention system. Storage system for liquid and solid wastes generated by the uranium refining and conversion process. This may include holding or settling ponds as well as approved shallow land burial sites.

LIST OF PARTICIPANTS

CANADA

Frost, S.E. Eldorado Nuclear Limited
Suite 400, Albert Street 255
Ottawa, Ontario K1P 6A9

FRANCE

Maqueron, Services Techniques de Protection
CEN de Fontenay-Aux-Roses
B.P. No. 6
92260 Fontenay-Aux-Roses

UNITED KINGDOM

Hayes, M.R. British Nuclear Fuels Ltd.
Health Physics and Safety Division
Springfield Works
Salwick, Preson, Lancs.

UNITED STATES OF AMERICA

Wing, J.F. Environmental Protection Branch
(Chairman) Oak Ridge Operations Office
P.O. Box E
Tennessee 37830

OBSERVERS

FRANCE

Peyran, P. COMURHEX
Rue Montceau - Paris

UNITED KINGDOM

Eccles, H. British Nuclear Fuels Ltd.
Springfield Works
Salwick, Preson
Lancs.

UNITED STATES OF AMERICA

Brown, B.E.

Kerr-McGee Nuclear Corporation
P.O. Box 25861
Oklahoma City, OK 73125

Bishop, J.C.

Allied Chemical Corporation
P.O. Box 8005 R
Morristown, NJ 07960

INTERNATIONAL ATOMIC ENERGY AGENCY (IAEA)

Zabaluev, Y.V.

(Scientific Secretary)

Division of Nuclear Safety and Env. Protection
P.O. Box 100, A-1400 Vienna