COMMERCIAL IN CONFIDENCE

AAEC/TM583

AAEC/TM583

# AUSTRALIAN ATOMIC ENERGY COMMISSION
## RESEARCH ESTABLISHMENT
## LUCAS HEIGHTS

# PRELIMINARY DESIGN AND COST CONSIDERATIONS FOR A PLANT TO PRODUCE NUCLEAR PURITY URANIUM DIOXIDE FROM AUSTRALIAN ORE CONCENTRATES

by

B.G. CHARLTON
P.G. ALFREDSON

March 1971

COMMERCIAL IN CONFIDENCE

AUSTRALIAN ATOMIC ENERGY COMMISSION
RESEARCH ESTABLISHMENT
LUCAS HEIGHTS

# PRELIMINARY DESIGN AND COST CONSIDERATIONS FOR A PLANT TO PRODUCE NUCLEAR PURITY URANIUM DIOXIDE FROM AUSTRALIAN ORE CONCENTRATES

by

B. G. CHARLTON
P. G. ALFREDSON

## ABSTRACT

Design considerations are outlined for plants for the production of nuclear purity uranium dioxide with capacities of 100, 200 and 500 tonnes U/year. The cost of the process equipment is not greatly affected by various process alternatives; equipment performance, which affects product quality, consistency of powder properties and plant reliability, is important in determining the recommended process. This involves the following steps: batch dissolution, continuous solvent extraction in mixer-settlers, single-stage precipitation of ADU, thickening and spray drying of ADU, and calcination-reduction in continuous pulsed fluidised bed reactors.

continued...

ABSTRACT (continued)

Estimates of the cost of recovery of free acid and combined nitrate from the raffinate and filtrate waste streams indicated that the value of the recovered acid would be greater than the processing cost only in the case of free acid recovery from solvent extraction raffinate in the 500 tonne/year plant. However if acid recovery is necessary for plant effluent control, the processing cost can be largely offset by the value of the recovered acid.

# CONTENTS

Page

continued...

CONTENTS (continued)

# 1. INTRODUCTION

This report outlines design considerations for plants to produce, from Australian ore concentrates, a ceramic grade uranium dioxide suitable for the fabrication of pellets for nuclear fuel elements. A standard process is described in detail and various alternative equipment units are compared on the basis of performance and cost data to identify cost sensitive areas and derive a recommended process.

The study is based on information obtained from the research programme at Lucas Heights (Alfredson 1970) and from the literature. Plant capacities of 100, 200 and 500 tonnes of uranium per year as uranium dioxide are considered. These throughputs were chosen to include the size of plant which, though not completely economic might be built to meet the early requirements of a power programme, and a larger plant which would be economic in a nuclear power programme. In the Appendix, estimates are also given for the capital cost of the installed process equipment only, and the annual direct processing charges excluding labour. These estimates are compared with other published data (Amdel 1966, 1970).

The standard process has the following stages:

(a) Batch dissolution
(b) Continuous solvent extraction in mixer-settlers
(c) Continuous single-stage precipitation
(d) Continuous rotary drum vacuum filtration
(e) Continuous belt drying
(f) Continuous pulsed bed calcination-reduction.

Acid is not recovered; acidic waste solutions are neutralised and mixed with alkaline wastes and are probably subjected to further waste treatment as required by local regulations; the latter is not included in this report.

The following alternatives are considered:

(a) Continuous dissolution
(b) Continuous pulsed column solvent extraction
(c) Continuous two-stage precipitation
(d) Continuous spray drying
(e) Continuous calcination-reduction in rotary kiln furnaces
(f) Batch-tray calcination-reduction.

The costs of recovery of free nitric acid from the raffinate, and the recovery as nitric acid of the combined nitrate contained in the raffinate and filtrate are also considered.

## 2. DESIGN CONSIDERATIONS FOR A PLANT USING THE STANDARD FLOWSHEET

The mass balance for the standard flowsheet is given in Figure 1 for a throughput of 500 tonnes U/yr. The mass balances for other throughputs are proportional to this. Figure 2 gives the equipment flowsheet.

### 2.1 Yellow Cake Specification, Storage and Handling

There is no standard specification for Australian yellow cake, but most modern treatment plant processes produce a high quality concentrate with the $U_3O_8$ content better than 90 per cent. Typical analyses for Rum Jungle concentrates held at Lucas Heights are as follows:

| $U_3O_8$ | $H_2O$ | $Na_2O$ | $SO_3$ | $SiO_2$ and Acid Insolubles |
|---|---|---|---|---|
| 92% | 1.0% | 3.5% | 0.5% | 0.25% |

While variations in Rum Jungle concentrates can be anticipated, they should cause no difficulties in subsequent processing. The solvent extraction purification process is very flexible and can cope with a wide variety of concentrates. The only concentrates which may be difficult to purify are those containing high concentrations of rare earth elements such as Mary Kathleen concentrates. Silica can also cause problems as it precipitates in acid solutions and hence can cause blockages in plant equipment, but the concentration in the Rum Jungle concentrates is well below that at which this trouble is likely to occur (approx. 1 per cent).

Yellow cake is contained in 44 gal steel drums with a polythene liner, holding about 600 lb of concentrate. The drums of yellow cake are safe to handle and since the $\gamma$ radiation dose on the surface of the drum is about 2.5 mrem/hr no special radiation precautions are necessary.

In the proposed plant, the drums are emptied into the dissolver from a ventilated dust hood. The plastic liner is disposed of as solid waste, and the empty drum monitored inside and out for contamination. If contaminated, it is washed clean for re-use. The contamination level should be less than 1 mrem/hr of fixed activity before despatch from the plant. The wash water can be used for make-up in the dissolver.

In general, the concentrate flows readily and does not pick up moisture in storage, provided the drum is not left open for any appreciable time. Normal safety procedures in operating the plant should minimise this occurrence.

### 2.2 Dissolution

Batch dissolution is proposed since it provides a convenient point for analysis and quantitative checking of the uranium input to the plant.

The batch dissolution equipment is designed on the basis of using two dissolver tanks, each capable of producing the uranyl nitrate required for approximately one shift of operation using a discrete number of drums of yellow cake, and allowing approximately 8 hours' residence time in the dissolver.

The contents of a drum of yellow cake are dumped in the dissolver and the calculated volume of nitric acid added from a head tank. The dissolver is stirred and heated with steam to near boiling (110°C). Water is added to make up the required volume. The solution is maintained at approximately 95°C until required, and analysed and adjusted for uranium and acid concentrations before use. A water-cooled jacketed pipe is provided to cool the solution to 30°C as it is fed to mixer-settler units.

Acid requirements for dissolution are based on experience at Lucas Heights which indicates that 1.35 litres of 16 M acid per kilogram of uranium (Charlton, A.A.E.C. unpublished work) is required to produce a 300 g U/$\ell$ feed solution with 3 M free acid concentration.

In overseas practice, the need to filter the dissolver solution before solvent extraction is approached in different ways. Springfields (U.K.A.E.A.) practice is to filter all solutions on a precoated rotary drum vacuum filter (Littlechild 1967). French practice is to decant the clear liquor and filter the settled liquors on a rotary drum vacuum filter (Decrop et al. 1958). Other producers in the United States (Thayer 1958) and Canada (Burger and Jardine 1958) do not filter this solution. We also use the solution without filtration (Charlton, A.A.E.C. unpublished work).

The Springfields process is fully reported (Mason and Smith 1965) and is based on concentrates containing 0.7 to 4.0 per cent silica which are blended to produce a solution of 1.8 per cent silica on a uranium basis. Rum Jungle concentrates which we have used contain approximately 0.25 per cent silica on a uranium basis. No troubles have been encountered with mixer-settlers due to this silica in the feed, though some blockage of the control valve and flowmeter used to control the feed rate has occurred. The feasibility of using a flowmeter and needle valve to control the flow required for a 200 tonnes/yr plant has been tested and no blockage or control problems were encountered. It is therefore assumed that it is not necessary to filter the dissolved yellow cake before solvent extraction.

### 2.3 Solvent Extraction

Uranyl nitrate is purified using 20 vol. per cent tributyl phosphate (TBP) in kerosene according to the flowsheet shown in Figure 3 in mixer-settler units similar to those currently in use at Lucas Heights. These units use 6 extraction stages, 10 scrub stages and 16 strip stages in 2 x 16-stage units (Charlton A.A.E.C unpublished work).

The Lucas Heights units are capable of processing 15 tonnes U/yr, which compares favourably with reported throughputs and sizes of the Springfields plant (Page et al. 1960). For this study, the Lucas Heights units have been scaled up on the basis of interfacial surface area, keeping the same length-breadth ratio and a residence time in the mixer of greater than one minute. A minimum residence time of 10 seconds is needed for at least 90 per cent efficiency of mass transfer (Lowes and Larkin 1967).

The proposed unit is controlled manually using flowmeters and valves on the feed streams. Uranium concentrations are measured continuously on the product stream and in the solvent in a critical extraction stage as described by Littlechild (1967). Gamma absorptiometry (Yates and May 1970) is recommended for this analysis. A frequent check is also made on the raffinate and recycle solvent streams to ensure that the uranium is not being carried through in these streams. The solvent extraction unit can be shut down by stopping all process streams, shutting down the mixers and closing the raffinate and product outlets, and can be restarted after a delay of several days with no significant loss of product quality.

### 2.3.1 Solvent washing

Littlechild (1967) reported that efficient carbonate washing is essential to prevent the concentration of uranium building up in the solvent. He recommended the use of five stages of carbonate washing, followed by a water (very dilute acid) wash to remove carbonate haze. He did not state the required ratio of carbonate to organic for the cleanup cycle, but Burger and Jardine (1958) reported the use of a ratio of 1:25 for pulse columns, and this is also assumed in this study.

### 2.3.2 Solvent loss and make-up

TBP will be lost to the raffinate and product streams because of its solubility in water (0.39 g/ℓ). It is assumed that the loss of TBP by evaporation is negligible.

Kerosene loss is more difficult to estimate. It is insoluble in water, but the evaporation rate is higher than for TBP. Evaporation losses should be minimised by the use of covered mixer-settler boxes and tanks. It has been arbitrarily assumed in this study that the kerosene loss is double that of TBP.

The concentration of TBP in the extractant is not critical and need only be checked on a weekly basis. It should be kept to 20 ± 1 per cent by TBP or kerosene addition as necessary.

### 2.3.3 Raffinate waste treatment and disposal

With six stages of extraction, the uranium concentration in the raffinate should be below 0.04 mg/ℓ. The acid concentration is 2.2 M and this is neutralised with lime before the raffinate is discharged to the effluent treatment plant. Three raffinate storage tanks are provided, each capable of holding 12 hours' throughput, to allow analysis and neutralisation to be carried out. They are operated on the basis of one filling, one being analysed, and one undergoing neutralisation and disposal.

### 2.3.4 Safety

A main reason for the choice of TBP as a solvent is its safety in handling. It is stable to high concentrations of nitric acid, and is not subject to spontaneous explosion reactions at operating conditions. The kerosene diluent is also chosen for its safety. It has a boiling range of 300-400°F, and a minimum flashpoint of 100°F. The mixture is generally handled in closed vessels and tanks and the fire risk is very low.

## 2.4 Precipitation

Continuous precipitation with ammonia gas is used in a single-stage, stirred tank reactor with a residence time of at least 8 minutes. A delay tank is provided between the precipitator and filter with a capacity equivalent to 6 hours' operation, holding 2 hours' supply normally, and allowing 4 hours in which to change the filter cloth in the event of a breakage. Both precipitator and delay tank are kept at 50°C by heating with steam.

The uranyl nitrate feed to the precipitator is controlled manually and the ammonia flow is automatically controlled to maintain the required pH (7.5) in the precipitator. Approximately 2.8 moles of ammonia per mole of uranium is required for this duty (Janov - A.A.E.C. unpublished work).

## 2.5 Filtration

The size of the required continuous rotary drum vacuum filter is calculated on the basis that an existing 3 $ft^2$ unit normally handles a slurry flow of 40 ℓ/hr, irrespective of wash water flow. The filter cloth specified is Eimco Filter Media No. NY-319F, this being the best of several types tested at Lucas Heights (Janov - A.A.E.C. unpublished work). This cloth has been in intermittent use for more than a year with no breakage or apparent deterioration but it is assumed that in plant service it may be necessary to change it every two months. The filter unit is easily operated, the flow of slurry being regulated to maintain a constant submergence of the drum.

The filtrate is fed to one of three delay tanks, one being filled, another being analysed and emptied, and a third available on standby. The concentration of dissolved uranium in the filtrate should be approximately 4 mg/ℓ, but the filtrate should also be checked for fine precipitate which may have carried through the filter cloth. This normally should only happen on start-up with a clean filter cloth.

Treatment of the filtrate depends upon how much uranium has passed through the filter. If no uranium precipitate is present, the liquid is discharged to the effluent treatment plant. If uranium precipitate is present, for example owing to a break in the filter cloth, it could be recycled via the third tank to be added slowly to the delay tank for re-filtering. In general, particles carried through the cloth under normal operation are usually extremely fine and do not settle on standing. Where neither of the above actions is appropriate, the filtrate may be treated with a flocculent to coagulate the particles which can then be recovered. No allowance for this treatment is included in this study.

The radioactivity of the alkaline filtrate is very low, being solely due to the uranium present. The concentration of ammonium nitrate is below 0.7 M and its recovery is not considered to be economic.

### 2.6 Drying

The wet cake from the filter contains approximately 30 wt. per cent moisture on a wet basis and must be dried before calcination-reduction. Continuous operation is desirable and three types of dryer have been considered, one being a spray dryer dealt with in Section 3.4. The other types considered are a conveyor-band (throughflow) dryer and a rotary dryer, for which the normal evaporation rates are 2 to 10 lb water/ft$^2$.hr and 2 to 6 lb water/ft$^3$.hr respectively. (Noden 1969). Drying rates for ammonium diuranate (ADU) have not been measured but no problems have been encountered in the use of batch ovens (Janov, A.A.E.C. unpublished work). The minimum drying rates for the two types of dryer considered above have been used to give conservative estimates for the cost of this equipment. On this basis, the conveyor-band dryer is 10-15 per cent cheaper than a rotary dryer and has been selected for this study.

The dryer is fed directly from the filter and the product is discharged into a hopper.

### 2.7 Calcination-Reduction

Various types of equipment are available for calcination-reduction of ADU to $UO_2$. In the standard flowsheet, a continuous pulsed bed is used, but rotary kiln furnaces and batch tray furnaces are also considered in Sections 3.5 and 3.6 respectively.

Fluidised-bed processes are commonly used in uranium processing (Harrington and Ruehle 1959, Alexander et al. 1960, Alfredson 1967) but difficulties arise in fluidising the high surface area, fine powders required for $UO_2$ pellet manufacture due to channeling of the gas through the bed and the elutriation of fine particles. A pulsed bed is being developed at Lucas Heights to overcome these problems (Alfredson 1970) and the proposed continuous pulsed bed unit is based on current experience with batch operation and projected modes of continuous operation (Fane and Alfredson 1970). The advantages of the pulsed fluidised bed reactor are good gas-solids contacting and heat transfer characteristics which should minimise microsintering of the powder during reduction and give uniform powder properties.

The ADU is calcined and reduced continuously in one pulsed bed heated to approximately 600°C, the $UO_2$ product being stabilised continuously at ambient temperature in a second pulsed bed. Dried and granulated ADU is fed into the top of the first pulsed bed by means of a screw feeder and rotary gate valve, powder is withdrawn from the bottom of the bed by a pipe, fitted with a slide valve, and fed by a cooled vibratory feeder into a cooled transfer hopper where it is fluidised with nitrogen. Similar feed and discharge mechanisms are provided for the stabilisation unit.

Automatic control of the bed temperature and level, continuous recording of the bed and filter pressure drops, and control of the pulsation by solenoid valves and a timer, is provided for in the reduction reactor. Similar controls, but without the temperature control, are provided for in the stabilisation reactor.

The size of the unit is based on an estimate that the 5 in diameter reactor in use at Lucas Heights can process approximately 10 kg/hr of uranium dioxide. The same length/diameter ratio for the bed and solids residence time are used. For mass balance calculations, gas requirements are assumed to be twice the stoichiometric hydrogen requirement for the reduction reaction and twice the stoichiometric oxygen requirement for the reaction $UO_2 \rightarrow UO_{2.1}$ for stabilisation, although the requirement of a constant fluidising gas velocity actually leads to varying stoichiometric requirements for different size equipment (Fane and Alfredson 1970).

Cracked ammonia is used for reduction without dilution at a flow equivalent to twice the stoichiometric requirement, and 2 per cent oxygen (10 per cent air) in nitrogen is used for stabilisation at the same volumetric flowrate as in the reduction reactor. Small amounts of inert gas are also required for purging hoppers, feeders, etc. but this requirement is assumed to be negligible.

Crushing of the dried ADU to approximately -20BSS is required and this is done in a Jackson and Crockett No. 6 granulator. From current experience, it is estimated that this granulator can process approximately 35 kg/hr ADU, equivalent to approximately 180 tonnes U/yr.

### 2.8 Utilities

In this section, utilities specifically concerned with process equipment are considered in some detail and those which would be shared in an integrated fuel manufacturing plant are briefly mentioned for completeness.

Fume control from the dissolver, dryer and calcination-reduction unit is by means of a sodium carbonate jet scrubber which can create a flow of 1,000 cfm air complete with the necessary ducting, circulation pump and storage tank. The gas from the reduction reactor will contain a maximum of 75 per cent hydrogen, which is explosive in air between 4 and 74 per cent concentration (Kirk-Othmer 1966). It is therefore diluted at least 30 times with air as soon as possible after passing through the reactor and then piped to the scrubber.

Yellow cake drums are tipped and ADU granulated in separate forced draught dust hoods, the air being filtered before discharge. General ventilation, filtration of exhaust air and a stack are not considered further in this study.

Nitric acid (60 per cent commercial grade) is delivered in multiples of 10 tons (1,600 gal) lots. It is stored in a ground level tank which can hold at least one week's supply, with a process head tank capable of holding 12 hours' supply.

Ammonia is supplied in 850 lb cylinders (no storage capacity required in the plant) for the 100 and 200 tonnes/yr plants and in 2 ton lots for the 500 tonnes/yr plant.

Demineralised water is standard for processing use throughout the plant, since a very high purity product is required. It is assumed that process water is obtained direct from the town supply.

Hydrogen is supplied by cracking ammonia over a heated catalyst and nitrogen is supplied by burning cracked ammonia in air in an inert gas generator. The commercial unit size of generators available is approximately 1000 $ft^3$/hr. Compressors and storage receivers are provided to supply the hydrogen and nitrogen to the pulsed fluidised beds at 50 psig. The compressed air requirement for the plant is small (5 $ft^3$/min for a 500 tonnes/yr plant) and is not considered further.

Electrical supply is usually installed in units of 200 kVA and it is assumed that one such unit will be sufficient for all sizes of plant considered in this study.

## 3. EQUIPMENT ALTERNATIVES

### 3.1 Continuous Dissolution

Continuous operation of a process is usually more attractive than batch operation since automatic control can be used, smaller equipment is required for a given throughput, and less manual handling is required. These advantages hold for continuous dissolution. However, a feed hopper and screw feeders are required with the continuous dissolver, but not with batch dissolvers. Batch operation also has the advantage that a convenient point is provided for uranium accounting. Operational requirements are much the same for batch and continuous dissolution.

Continuous dissolution has been thoroughly investigated at Springfields (Page et al. 1960) where it was recommended that a three-stage system with a total residence time of 8 hours be adopted. Three equal-volume stirred stages are proposed for this study, most of the reaction taking place in the first stage, to which yellow cake, nitric acid and water are fed continuously, the temperature being maintained at near boiling temperature (about 110°C) by heating with steam. The second and third stages are maintained at about 95°C. The residence time allowed permits the use of the third stage as a feed tank to the solvent extraction process. The dissolver could be shut down and restarted without significantly affecting the concentration of the product.

The dissolver product solution should be analysed for uranium and free acid. Uranium may be analysed continuously using gamma absorptiometry (Yates and May 1970). The residence time in the final stage is long enough for sampling every 2 hours to be sufficient.

### 3.2 Pulsed Column Solvent Extraction

Pulsed columns for continuous processing are an alternative to mixer-settlers and have the advantage of a smaller hold-up. Thus they reach equilibrium much quicker and are more responsive to control than mixer-settlers. They are also easier to clean. Whereas mixer-settlers are often fed by gravity, pulsed columns are much taller and usually require continuously operating feed pumps. An efficient pulsing unit is also necessary and interface control can cause problems, particularly if large quantities of interfacial crud are formed.

Conditions of flow required are very similar to those examined by Logsdail and Larner (1963) and calculations are based on the optimum frequency conditions established by them.

The sizes of the pulsed column units required are as follows:

| | | 100 tonnes/yr | 200 tonnes/yr | 500 tonnes/yr |
|---|---|---|---|---|
| Extraction and Scrub: | diameter | 3 in | 4 in | 6 in |
| | height | 26 ft | 26 ft | 26 ft |
| Strip : | diameter | 6 in | 8 in | 12 in |
| | height | 31 ft | 31 ft | 31 ft |

Additional height is required in the process building to accommodate the pulsed columns, but a smaller floor area is required than for mixer-settlers. These considerations have been neglected in comparing the costs of these alternatives.

### 3.3 Two-Stage Precipitation

The advantage of two-stage or multi-stage precipitation over single-stage precipitation is that a wider range of ADU properties can be achieved and control of the unit should be less critical. The first stage of precipitation is carried out at a pH where only part of the uranium is precipitated and small changes in operating conditions are not important. The pH in the second stage is maintained in the range 7 to 9 and does not need to be automatic.

The precipitation conditions necessary to yield a satisfactory ADU for pellet manufacture produce a precipitate which filters at much the same rate as single state precipitated ADU (Janov et al. 1970).

The equipment used is similar to that for single-stage precipitation; two stirred tanks of equal volume are used with a total residence time of 16 minutes. Purified uranyl nitrate is fed to the first stage where ammonia is added continuously to precipitate the bulk of the uranium as ADU. The slurry then overflows to the second stage where a further addition of ammonia is made to bring the slurry to the required pH. It then overflows into the filter delay tank.

### 3.4 Spray Drying

Spray drying gives control over the particle size of the dry ADU and eliminates the need for crushing before pulsed bed calcination and reduction. No detrimental effects of spray drying on the properties of the $UO_2$ powder have been observed (Levins, A.A.E.C. unpublished work).

Spray drying can be substituted for three processing stages in the standard flowsheet; filtration, drying and granulation, provided that a counter-current decantation stage is included to give as concentrated a slurry feed as possible to the dryer and to eliminate ammonium nitrate from the solution. The cost of the dryer depends upon the rate of water evaporation required. Its physical size determines the maximum particle size of powder obtainable.

The settling rate for single-stage precipitated (pH 7.5) ADU is approximately 1.2 ft/hr and the decanter size is calculated by assuming that the required feed to the dryer is 35 wt. per cent solids.

### 3.5 Calcination-Reduction in Rotary Kiln Furnaces

Rotary kiln furnaces have been used for the production of sinterable $UO_2$ in France (Delange 1963), Germany (Wirths and Ziehl 1958) and England (Smith et al. 1964). They have the advantage of continuous and simple operation but require a closely sized feed material.

Separate calcination, reduction and stabilisation reactors have been assumed with respective mean residence times of 20 min, 1 hour, and 20 min. Delmas and Holder (1963) reported a throughput of 72 tonnes/yr for reduction in a 6 inch diameter by 12 ft long furnace, but a commercial quotation received at Lucas Heights indicated three times this flow. The sizes of the reduction units required have been calculated by scaling the diameter allowing for a maximum available diameter of 10 in and assuming a constant length. For calcination and stabilisation, it is assumed that a reduction in residence time can yield a proportionate increase in throughput as controls of speed and slope are available in these units and the diameters are scaled accordingly. The resultant dimensions have been checked for consistency with the controlling variables of speed and slope by use of the equation of Sullivan et al. (1927):

$$\bar{t} = 0.19\ L/ND \sin \alpha \quad ,$$

where $\bar{t}$ is the mean residence time in a kiln of diameter D and length L rotating at a speed N and set at an inclination $\alpha$ to the horizontal.

Ancillary equipment required includes transfer hoppers from the dryer, screw feeder and hopper from the reduction reactor, a cooled screw feeder between the reduction and stabilising reactors and product containers for transporting the $UO_2$ from the plant. Rotary kiln furnaces will require much more floor space than the pulsed fluidised bed reactors and about the same as the batch tray furnaces, but no allowance is made for this in the comparison of costs.

### 3.6 Batch Tray Calcination-Reduction

This type of unit has been used in Canada (Berry 1967), United States (Placek and North 1960) and England (Hinton 1955). It is at present in use at Lucas Heights for producing small quantities of $UO_2$ (5 kg batches) and is known to be capable of producing satisfactory $UO_2$ (Janov - A.A.E.C. unpublished work). It has the disadvantages of requiring much manual handling in loading and unloading the furnace, the possibility of non-uniform temperatures occurring within the bed of powder, and possible differences in powder characteristics between batches.

For the proposed plant, a continuously operating furnace is assumed. The ADU powder is loaded into trays to be stacked in a reaction vessel which is then sealed, purged and placed in the furnace for reaction. After reaction, the vessel is removed and placed to one side to cool; meanwhile another vessel has been loaded and replaces the first one in the furnace. After the vessel and contents have cooled, the powder is stabilised and then unloaded. The full cycle for each vessel takes 12 hours, as follows: 2 hours' discharge and load; 2 hours' heat up and reaction; 6 hours' cooling and 2 hours' stabilisation. On this basis, the furnace could accommodate 6 reaction vessels in 12 loadings per day, but a total of 5 vessels with 10 loadings per day is assumed for this plant.

The volume of the furnace and vessels is calculated by assuming that the bed of powder in the trays is one inch deep, there is 2 inches between each tray and the bulk density of ADU is 62.4 $lb/ft^3$.

Control of the furnace temperature is automatic while gas flow rates are controlled manually with flowmeters. The heating load for all types of calcination-reduction apparatus is assumed to be similar.

## 4. NITRIC ACID RECOVERY

Approximately 16 per cent of the nitric acid requirement for the solvent extraction flowsheet is used in the neutralisation of the sodium in the yellow cake and becomes sodium nitrate in the raffinate, 46 per cent appears as free acid in the raffinate and 38 per cent appears as ammonium nitrate in the filtrate. Since nitric acid accounts for approximately 60 per cent of the cost of chemicals for conversion of yellow cake to $UO_2$ powder by the standard flowsheet, there is an economic incentive for acid recovery. Depending on the location of the processing plant, there may be limits on the nitrate content of the plant effluent and this could necessitate removal of nitrate from the effluent stream. Recovery of this nitrate in the form of nitric acid would give some economic return for this processing.

Two alternative approaches for nitric acid recovery are considered:

(i) recovery of free acid from the raffinate solution only, and
(ii) recovery from nitrate in raffinate and filtrate solutions.

Recovery of free nitric acid from raffinate streams at the National Lead Company of Ohio has been described by Harrington and Ruehle (1959) and some details of the evaporation and distillation of these streams are also available (Arnold et al. 1956). For this study, it is assumed that 90 per cent of the raffinate is boiled off in a stainless steel forced-circulation evaporator and the vapours distilled in a bubble-cap tower, fitted with a steam-heated external reboiler and a surface condenser. The dimensions of the distillation tower were calculated by assuming a superficial vapour velocity of 0.6 ft/sec, 20 bubble-cap plates at 24 in spacing, and an operational pressure of 7 psia. Allowance is also made for ozonisation of the liquid on one plate to oxidise chlorine and minimise corrosion effects (Arnold et al. 1956). The design of the evaporator is based on a feed stream of 2.2 M free nitric acid with 90 per cent evaporation at 2.8 psia, the overall heat transfer coefficient being 250 Btu/hr.ft$^2$.°F with a steam supply of 50 psig.

Nitrate can be recovered from raffinate and filtrate by the addition of sulphuric acid equivalent to the combined nitrates present in both liquids and boiling off the free nitric acid formed. This type of process is used at Port Hope Refinery, Canada, on raffinate streams from pulsed column operations (Burger and Jardine 1958). The sizes of evaporator and distillation column necessary to handle the greatly increased volume of liquid have been calculated on the same basis as for acid recovery from the raffinate stream alone.

## 5. BASIS FOR COSTING

This Section outlines the basis for the cost estimates made in the comparison of the flowsheet alternatives described above and in the Appendix.

Equipment costs were determined from published data (Aries and Newton 1955; Bauman 1964; Buchanan and Sinclair 1964; Drew and Ginder 1970; Guthrie 1969a, 1969b; Perry 1963; Peters and Timmerhaus 1968). In general, the size of the plant is relatively small for chemical plant, and in many cases it is difficult to find reasonable cost figures. When the equipment size required is below that quoted in the literature, the cost has been calculated by extrapolation with due regard to the type of equipment. Where several references have yielded different cost figures, a reasonable mean has been taken. Except where otherwise stated, type 316 stainless steel has been assumed to be the material of construction of the equipment.

Particular difficulty was encountered in arriving at reasonable estimates for the mixer-settlers, pulsed solvent extraction column, pulsed bed calcination-reduction units and rotary kiln furnaces. Costs for these are not directly obtainable from the literature. Costs were estimated for the first three on the basis of material, parts and manufacturing costs from a knowledge of the equipment in use at Lucas Heights (for mixer-settlers and pulsed beds) and that used at Harwell (for solvent extraction columns). The costs of rotary calcining furnaces used in this study are based on commercial quotations received at Lucas Heights for units approaching 100 tonnes/yr throughput.

All capital costs for equipment have been updated by the use of the Marshall and Stevens installed equipment index (published bi-monthly in Chemical Engineering) to January 1970.

The costs of process chemicals, except nitric acid, were taken from Australian Chemical Process Engineering for June 1970; this listing is published quarterly. Costs of nitric acid were obtained from George Shirley Pty. Ltd., Sydney, on the basis of supply in bulk of at least 100 tons/yr of 60 per cent acid which is the most economical concentration to purchase.

## 6. DISCUSSION

### 6.1 Equipment Alternatives

Details of the costs of alternative equipment to that used in the standard flowsheet are given in Table 1 and summarised in Table 2. The estimated costs for continuous dissolution are slightly greater than for batch dissolution and, as a uranium accounting point is also lost, the use of continuous dissolution would not seem to be justified for the sizes of plant under consideration. Pulsed columns appear to be slightly cheaper for comparable throughputs than mixer-settlers although, with the degree of uncertainty in the assumptions used in estimating these costs, actual costs could be more comparable. Two-stage precipitation increases the equipment cost slightly but this may be compensated by the need for slightly less stringent control in precipitation.

Some economic advantage is obtainable by the use of spray drying, in which two process stages replace three, and manpower requirements are also probably reduced. The difference between heating requirements for the two processing routes is considered to be small. The calculated costs for rotary kiln furnaces are much greater than for continuous pulsed bed equipment even allowing for the greater uncertainty of the cost data for the latter. Batch-tray furnaces are roughly comparable in cost to pulsed bed equipment, but the manpower requirements are considered to be greater.

Thus, with one exception, the cost of the process equipment is not greatly affected by the various alternatives to the standard flowsheet considered. Equipment performance which affects product quality, consistency of powder properties and plant reliability and has been described in the previous Sections is therefore important in determining which process is to be recommended. This involves the following steps: batch dissolution, continuous solvent extraction in mixer-settlers, continuous single-stage precipitation, continuous counter-current decantation and thickening, spray drying, and continuous calcination-reduction in pulsed fluidised bed reactors.

### 6.2 Nitric Acid Recovery

The costs of recovering nitric acid from raffinate produced by the standard flowsheet are detailed in Table 3 and summarised in Table 4. These estimates do not include engineering design, construction and startup costs in the capital costs, nor capital charges in the operating costs. It appears to be uneconomic to recover the free acid and nitrate as acid from filtrate and raffinate solutions since the processing cost (excluding capital charges) is greater than the value of the recovered acid. However, recovery of the free acid only from the raffinate in the 500 tonnes/yr plant is attractive. If limits on nitrate in the plant effluent make processing essential, a large proportion of this processing cost can be offset by the reduced acid requirement brought about by using this recovery system.

## 7. SUMMARY

Design considerations have been presented for the production from Australian ore concentrates of ceramic grade uranium dioxide in plants with capacities of 100, 200 and 500 tonnes of contained uranium per year. The standard process is described and compared with various flowsheet alternatives on the basis of performance and capital and operating costs.

The cost of the process equipment is not greatly affected by the various alternatives and the more important factor in determining the preferred process is equipment performance which affects product quality, consistency of powder properties and plant reliability.

The recommended process has the following steps: batch dissolution, continuous solvent extraction in mixer-settlers, continuous single-stage precipitation, continuous counter-current decantation and thickening, spray drying, and continuous pulsed bed calcination, reduction and stabilisation.

Estimates of the costs of recovery of free nitric acid from raffinate and of nitric acid from the combined nitrate in the raffinate and filtrate streams indicate that it is marginally economic to recover free nitric acid from the raffinate stream in the largest plant considered, but in no other case is recovery economic. However to make the plant effluent meet acceptable nitrate concentrations, recovery may be necessary and this processing cost can be largely offset by the value of the recovered acid.

## 8. ACKNOWLEDGEMENT

The assistance of members of the Chemical Engineering Section in the preparation of this report is gratefully acknowledged as well as useful discussions with Messrs. W. J. Wright and J. M. Silver of Nuclear Development Division.

## 9. REFERENCES

Alexander, R. C., Shortis, L. P. and Turner, C. J. (1960). - The Second Uranium Plant at Springfields. Trans. Instn. Chem. Engrs. 38, 177-183.

Alfredson, P. G. (1967). - The application of fluidised bed processes in nuclear chemical engineering. Paper presented at Fluidisation Symposium, Monash University, September 1967.

Alfredson, P. G. (1970). - The production of nuclear grade uranium dioxide from Australian ores. Proc. Aus. I.M.M. No. 233, 61-68. Also AAEC/TM514.

Amdel (1966). - Cost estimate for production of ceramic grade $UO_2$ in Australia. Amdel Report No. 462.

Amdel (1970). - Cost estimate for production of nuclear grade $UO_2$ in Australia. Amdel 2/6/13 Progress Report No.1, March to September 1970.

Aries, R. S. and Newton, R. D. (1955). - Chemical Engineering Cost Estimation. McGraw Hill, N.Y.

Arnold, D. S., Whitman, A. and Podlipec, F. J. (1956). - Nitric acid recovery from raffinate by evaporation and fractional distillation. Chem. Eng. Progr. 52(9):362.

Bauman, H. C. (1964). - Fundamentals of Cost Engineering in the Chemical Industry. Reinhold, N.Y.

Berry, R. M. (1967). - Ceramic oxide production at Eldorado. Canadian Nuclear Technology, 6(2):30-33.

Buchanan, R. H. and Sinclair, C. G. (1964). - Cost and Economics of the Australian Process Industries. West Publishing Corporation Pty. Ltd., Sydney.

Burger, J. C. and Jardine, J. McN. (1958). - Canadian refining practice. Proc. 2nd Int. Conf. on Peaceful Uses of Atomic Energy, 4, 228.

Decrop, J., Delange, M., Holder, J., Huet, H., Sauteron, J. and Vertes, P. (1958). - Improvements in the purification of uranium compounds and in the production of uranium metal at the Bouchet factory. Proc. 2nd Int. Conf. on Peaceful Uses of Atomic Energy, 4, 10.

Delmas, R. and Holder, J. (1963). - Study of the preparation of sinterable uranium dioxide for the fabrication of fuel for EL 4. New Nuclear Materials, Including Non-metallic Fuels, Vol. 1, pp.95-108, IAEA, Vienna. (Translation: AAEC Report Lib/Trans. 189).

Delange, M. (1963). - The industrial production of $UO_2$ suitable for sintering at the Bouchet factory. New Nuclear Materials, Including Non-metallic Fuels. Vol. 1, p.65, IAEA Vienna. (Translation: AAEC Report Lib/Trans. 188).

Drew, J. W. and Ginder, A. F. (1970). - How to estimate the cost of pilot plant equipment. Chemical Engineering, 77(3), 9th Feb., p.100.

Fane, A. G. and Alfredson, P. G. (1970). - The production of sinterable uranium dioxide from ammonium diuranate in a pulsed fluidised bed reactor - interim report. AAEC/TM579.

Guthrie, K. M. (1969a). - Capital cost estimating. Chemical Engineering, 76(6), 24th March, p.114.

Guthrie, K. M. (1969b). - Costs (liquids handling). Chemical Engineering 76(8), 14th April, p.201.

Harrington, C. D. and Ruehle, A. E. (1959). - Uranium Production Technology. Van Nostrand, Princeton, N.J.

Hinton, C. (1955). - Some aspects of chemical processes ancillary to atomic energy - the manufacture of uranium metal from ore. Trans. Instn. Chem. Engrs. 33(1):45-51.

Kirk-Othmer, (A. Standen ed.) (1966). - Encylopaedia of Chemical Technology, 2nd Edition, Interscience, N.Y.

Janov, J., Alfredson, P. G. and Vilkaitis, V. K. (1970). - The influence of precipitation conditions on the properties of ammonium diuranate and uranium dioxide powder. AAEC Report (in press).

Littlechild, J. E. (1967). - Operational development of a uranium ore solution and extraction plant. I. Chem. E. Symp. Ser. No. 26, pp.107-110.

Logsdail, D. H. and Larner, G. S. (1963). - High throughputs in pulsed plate columns for uranium - tributyl phosphate system. AERE-R-4408.

Lowes, L. and Larkin, M. J. (1967). - Some design principles for large mixer-settlers. I. Chem. E. Symp. Ser. No. 26, pp.111-114.

Mason, T. and Smith, G. R. (1965). - Filtration of uranium extraction liquors. Filtration and Separation, July/August 1965, pp.278-286.

Noden, D. (1969). - Industrial driers - selection, sizing and costs. Chem. Process Eng. 50(10):67-76.

Page, H., Shortis, L. P. and Dukes, J. A. (1960). - The processing of uranium ore concentrates and recycle residues to purified uranyl nitrate solution at Springfields. Trans. Instn. Chem. Engrs., 38, 184.

Perry, J. H. (ed.) (1963). - Chemical Engineers Handbook. McGraw Hill, N.Y.

Peters, M. S. and Timmerhaus, K. D. (1968). - Plant Design and Economics for Chemical Engineers, McGraw Hill, N.Y.

Placek, C. and North, E. D. (1960). - Uranium dioxide nuclear fuel. Ind. Eng. Chem. 52(6):458-464.

Smith, S. E., Broadley, J. S., Brown, F. and Kent, W. C. L. (1964). - Development of large-scale manufacturing processes and plant for plutonium fast reactor fuel. Proc. 3rd U.N. Conf. on Peaceful Uses of Atomic Energy, 10, 161-167.

Sullivan, J. D., Maier, C. G. and Ralston, O. C. (1927). - U.S. Bureau of Mines, Technical Paper No. 384.

Thayer, H. E. (1958). - The newest United States uranium processing plant. Proc. 2nd Int. Conf. on Peaceful Uses of Atomic Energy, 4 22.

Wirths, G. and Ziehl, L. (1958). - Special problems connected with the production of uranium metal and uranium compounds. Proc. 2nd Int. Conf. on Peaceful Uses of Atomic Energy, 4, 16.

Yates, P. B. and May, J. R. (1970). - Analysis of uranium solutions by gamma absorptiometry. AAEC/TM557.

TABLE 1

DETAILED COSTS OF FLOWSHEET OPTIONS

| Item | 100 Tonnes/Yr |  | 200 Tonnes/Yr |  | 500 Tonnes/Yr |  |
|---|---|---|---|---|---|---|
|  | Unit Size | Cost, $A | Unit Size | Cost, $A | Unit Size | Cost, $A |
| 1a. Batch Dissolution |  |  |  |  |  |  |
| Tanks, Heated and Agitated, 2 off | 200 gal | 3,000 | 200 gal | 3,000 | 500 gal | 5,000 |
| Level Measurement |  | 250 |  | 250 |  | 250 |
| Temperature Measurement and Control |  | 1,450 |  | 1,450 |  | 1,450 |
| Nitric Acid Tank | 100 gal | 350 | 100 gal | 350 | 250 gal | 550 |
| Nitric Acid Pump | 10 gal/min | 450 | 10 gal/min | 450 | 25 gal/min | 600 |
| Nitric Acid Level Measurement and Pump Cut-Out |  | 300 |  | 300 |  | 300 |
|  |  | 5,800 |  | 5,800 |  | 8,150 |
| 1b Continuous Dissolution |  |  |  |  |  |  |
| Feeder | 40 lb/hr | 1,500 | 80 lb/hr | 2,000 | 200 lb/hr | 2,000 |
| Hopper | 10 $ft^3$ | 150 | 10 $ft^3$ | 150 | 30 $ft^3$ | 400 |
| Tanks, Heated and Agitated, 3 off | 35 gal | 2,100 | 70 gal | 3,000 | 150 gal | 3,900 |
| Control Equipment: Temperature recording and control Fluid Flow |  | 2,500 |  | 2,500 |  | 2,500 |
|  |  | 6,250 |  | 7,650 |  | 8,800 |
| 2a Mixer-Settler Solvent Extraction |  |  |  |  |  |  |
| Construction, Framework and Assembly |  | 10,000 |  | 12,000 |  | 16,000 |
| Agitators |  | 4,700 |  | 5,000 |  | 5,400 |
| Control Equipment and Panel |  | 4,400 |  | 4,400 |  | 4,500 |
|  |  | 19,100 |  | 21,400 |  | 25,900 |
| 2b Pulsed Column Solvent Extraction |  |  |  |  |  |  |
| Columns { Extraction Scrub | 3 in dia. x 26 ft | 1,800 | 4 in dia. x 26 ft | 2,000 | 6 in dia. x 26 ft | 3,000 |
| Columns { Strip | 6 in dia. x 31 ft | 2,800 | 8 in dia. x 31 ft | 5,500 | 12 in dia. x 31 ft | 8,500 |
| Plates |  | 2,000 |  | 2,900 |  | 3,500 |
| Pulsing Mechanisms |  | 3,500 |  | 3,500 |  | 3,500 |
| Control, Interface and Pulse Amplitude and Frequency |  | 1,000 |  | 1,000 |  | 1,000 |
|  |  | 11,100 |  | 14,900 |  | 19,500 |

continued...

TABLE 1 (continued)

| | Item | 100 Tonnes/Yr | | 200 Tonnes/Yr | | 500 Tonnes/Yr | |
|---|---|---|---|---|---|---|---|
| | | Unit Size | Cost, $A | Unit Size | Cost, $A | Unit Size | Cost, $A |
| 3a | Single-Stage Precipitation | | | | | | |
| | Precipitation Vessel (agitated) | 12 gal | 300 | 25 gal | 350 | 65 gal | 500 |
| | Control Equipment (pH, temperature, recording) | | 2,000 | | 2,000 | | 2,000 |
| | | | 2,300 | | 2,350 | | 2,500 |
| 3b | Two-Stage Precipitation | | | | | | |
| | Agitated Vessels, 2 off | 12 gal | 600 | 25 gal | 700 | 65 gal | 900 |
| | Control Equipment | | 2,200 | | 2,200 | | 2,200 |
| | | | 2,800 | | 2,900 | | 3,100 |
| 4a | Filtration, Drying and Granulation | | | | | | |
| | Delay Tank (agitated) | 200 gal | 750 | 350 gal | 1,000 | 1,000 gal | 1,800 |
| | Mono Pump | 50 gal/hr | 300 | 100 gal/hr | 300 | 250 gal/hr | 350 |
| | Temperature Control | | 200 | | 200 | | 200 |
| | Rotary Drum Vacuum Filter | 12 $ft^2$ | 13,500 | 25 $ft^2$ | 20,000 | 54 $ft^2$ | 24,000 |
| | Filtrate Tanks, 2 off | 400 gal | 1,500 | 1,000 gal | 2,800 | 2,000 gal | 4,450 |
| | Filtrate Discharge Pump | 20 gal/min | 500 | 50 gal/min | 600 | 100 gal/min | 700 |
| | Dryer Unit (including roller extruder) | 10 $ft^2$ | 20,000 | 20 $ft^2$ | 21,500 | 50 $ft^2$ | 26,000 |
| | Transfer Hoppers, 2 off | 3 $ft^3$ | 500 | 5 $ft^3$ | 600 | 12 $ft^3$ | 700 |
| | Jackson-Crockett Granulators, (35 kg/hr) | 1 No. | 1,300 | 2 No. | 2,600 | 3 No. | 3,900 |
| | Glovebox | 40 $ft^3$ | 400 | 80 $ft^3$ | 500 | 120 $ft^3$ | 600 |
| | Transfer Hoppers, 2 off | 3 $ft^3$ | 450 | 5 $ft^3$ | 450 | 12 $ft^3$ | 600 |
| | | | 39,400 | | 50,550 | | 63,300 |
| 4b | Thickening - Spray Drying | | | | | | |
| | Spray Drying Unit | 80 lb/hr | 15,000 | 160 lb/hr | 26,000 | 400 lb/hr | 39,000 |
| | Off-Gas Treatment - Acid Jet Scrubber | 100 $ft^3$/min | 1,000 | 100 $ft^3$/min | 1,000 | 100 $ft^3$/min | 1,000 |
| | Thickener | 3 $ft^2$ | 850 | 6 $ft^2$ | 1,100 | 15 $ft^2$ | 1,950 |
| | | | 16,850 | | 28,100 | | 41,950 |

continued...

TABLE 1 (continued)

| Item | 100 Tonnes/Yr | | 200 Tonnes/Yr | | 500 Tonnes/Yr | |
|---|---|---|---|---|---|---|
| | Unit Size | Cost, $A | Unit Size | Cost, $A | Unit Size | Cost, $A |
| 5a Continuous Pulsed Bed Calcination-Reduction | | | | | | |
| Calcination-Reduction Unit - Construction and Assembly | 6 in dia. | 9,000 | 8 in dia. | 12,000 | 10 in dia. | 13,500 |
| Furnace | | 8,000 | | 9,000 | | 11,000 |
| Stabilisation Unit - Construction and Assembly | 6 in dia. | 8,000 | 8 in dia. | 10,000 | 10 in dia. | 12,500 |
| Controls and Panel | | 9,000 | | 9,000 | | 9,000 |
| | | 34,000 | | 40,000 | | 46,000 |
| 5b Continuous Rotary Calcination-Reduction | | | | | | |
| Reduction Reactor (including temperature recording and control) | 6 in dia. | 36,000 | 6 in dia. | 36,000 | 10 in dia. | 50,000 |
| Calcination Reactor | 4 in dia. | 28,000 | 6 in dia. | 36,000 | 10 in dia. | 50,000 |
| Stabilisation | 4 in dia. | 13,600 | 6 in dia. | 21,000 | 10 in dia. | 25,000 |
| Gas Supply Panel | | 4,200 | | 4,200 | | 4,200 |
| | | 81,800 | | 109,200 | | 129,200 |
| 5c Batch Tray Calcination-Reduction | | | | | | |
| Note: Granulation not required | | | | | | |
| Furnace | 4 $ft^3$ | 5,500 | 8 $ft^3$ | 6,500 | 20 $ft^3$ | 8,500 |
| Vessels, 5 off | | 6,500 | | 7,500 | | 12,500 |
| Trays | 50 No. | 750 | 100 No. | 1,250 | 200 No. | 2,600 |
| Powder Handling Box | 250 $ft^3$ | 750 | 250 $ft^3$ | 750 | 250 $ft^3$ | 750 |
| Dust Filter | 2,000 $ft^3$/min | 6,000 | 2,000 $ft^3$/min | 6,000 | 2,000 $ft^3$/min | 6,000 |
| Control, Temperature and Gas | | 3,500 | | 3,500 | | 3,500 |
| | | 23,000 | | 25,500 | | 33,850 |

# TABLE 2

## SUMMARY OF COSTS OF FLOWSHEET OPTIONS ($A)

| | | 100 Tonnes/Yr | 200 Tonnes/Yr | 500 Tonnes/Yr |
|---|---|---|---|---|
| 1a | Batch dissolution | 5,800 | 5,800 | 8,150 |
| 1b | Continuous dissolution | 6,250 | 7,650 | 8,800 |
| 2a | Mixer-settler solvent extraction | 19,100 | 21,400 | 25,900 |
| 2b | Pulsed column solvent extraction | 11,100 | 14,900 | 19,500 |
| 3a | Single-stage precipitation | 2,300 | 2,350 | 2,500 |
| 3b | Two-stage precipitation | 2,800 | 2,900 | 3,100 |
| 4a | Filtration, drying and granulation | 39,400 | 50,550 | 63,300 |
| 4b | Spray drying, including thickness | 16,850 | 28,100 | 41,950 |
| 5a | Continuous pulsed bed calcination-reduction | 34,000 | 40,000 | 46,000 |
| 5b | Continuous rotary kiln calcination-reduction | 81,000 | 109,200 | 129,200 |
| 5c | Batch tray calcination-reduction | 23,000 | 25,500 | 33,850 |

## TABLE 3

## NITRIC ACID RECOVERY COSTS

(A) Free Acid Recovery

| Capital Cost<br>Item | Unit Cost | 100 Tonnes/Yr<br>Unit Size | <br>Cost, $A | 200 Tonnes/Yr<br>Unit Size | <br>Cost, $A | 500 Tonnes/Yr<br>Unit Size | <br>Cost, $A |
|---|---|---|---|---|---|---|---|
| Evaporator |  | 5 ft$^2$ | 20,000 | 10 ft$^2$ | 24,000 | 25 ft$^2$ | 37,000 |
| Distillation Column |  | 8 in dia. | 14,000 | 12 in dia. | 17,000 | 18 in dia. | 21,000 |
| Additional Steam Installation (70% efficiency) | $7 lb/hr | 230 lb/hr | 1,500 | 460 lb/hr | 3,000 | 1,150 lb/hr | 8,000 |
|  |  |  | 35,500 |  | 44,000 |  | 66,000 |
| Installation Costs, (20% equipment cost) |  |  | 7,100 |  | 8,800 |  | 13,200 |
|  |  |  | 42,600 |  | 52,800 |  | 79,200 |
| **Operating Cost** |  | Requirement |  | Requirement |  | Requirement |  |
| Steam | $1/10$^6$ Btu | 1.6 x 10$^9$ Btu | 1,600 | 3.3 x 10$^9$ Btu | 3,300 | 8.3 x 10$^9$ Btu | 8,300 |
| Cooling Water | $0.20/10$^3$ gal | 4 x 10$^6$ gal | 800 | 8.25 x 10$^6$ gal | 1,650 | 2.08 x 10$^7$ gal | 4,000 |
| Maintenance, 10% Capital Cost |  |  | 4,200 |  | 5,300 |  | 7,900 |
| Operation, 1 man full time | $4,000 shift | 4 shifts | 16,000 | 4 shifts | 16,000 | 4 shifts | 16,000 |
|  |  |  | 22,600 |  | 26,250 |  | 36,200 |
| Value of Recovered Nitric Acid | $109/ton | 100 tons | 10,900 | 180 tons | 19,600 | 450 tons | 49,000 |

(B) Recovery of Both Nitrate and Free Acid

| Capital Cost<br>Item | Unit Cost | 100 Tonnes/Yr<br>Unit Size | <br>Cost, $A | 200 Tonnes/Yr<br>Unit Size | <br>Cost, $A | 500 Tonnes/Yr<br>Unit Size | <br>Cost, $A |
|---|---|---|---|---|---|---|---|
| Evaporator |  | 22 ft$^2$ | 34,000 | 44 ft$^2$ | 43,000 | 110 ft$^2$ | 64,000 |
| Distillation Column |  | 18 in dia. | 21,000 | 24 in dia. | 25,000 | 36 in dia. | 34,000 |
| Additional Steam Installation | $7 lb/hr | 900 lb hr | 6,300 | 1,800 lb/hr | 12,600 | 4,500 lb/hr | 31,500 |
|  |  |  | 61,300 |  | 80,600 |  | 129,500 |
| Installation Costs, (20% equipment cost) |  |  | 12,300 |  | 16,100 |  | 25,900 |
|  |  |  | 73,600 |  | 96,700 |  | 155,400 |
| **Operating Cost** |  | Requirement |  | Requirement |  | Requirement |  |
| Sulphuric Acid 98% Commercial | $45 ton | 66 tons | 3,000 | 132 tons | 6,000 | 330 tons | 14,900 |
| Steam | $1/10$^6$ Btu | 6.5 x 10$^9$ Btu | 6,500 | 1.3 x 10$^{10}$ Btu | 13,000 | 3.24 x 10$^{10}$ Btu | 32,400 |
| Cooling Water | $0.20/10$^3$ gal | 1.62 x 10$^7$ gal | 3,200 | 3.34 x 10$^7$ gal | 6,600 | 8.45 x 10$^7$ gal | 16,900 |
| Maintenance, 10% Capital Cost |  |  | 7,400 |  | 9,700 |  | 15,500 |
| Operation, 1 man full time | $4,000 shift | 4 shifts | 16,000 | 4 shifts | 16,000 | 4 shifts | 16,000 |
|  |  |  | 36,100 |  | 51,300 |  | 95,700 |
| Value of Recovered Nitric Acid | $109 ton | 212 tons | 23,100 | 425 tons | 46,300 | 1,010 tons | 110,000 |

# TABLE 4

## SUMMARY OF NITRIC ACID RECOVERY COSTS ($A)

| | Costs for Three Plant Throughputs (tonnes U/Yr) | | |
|---|---|---|---|
| | 100 | 200 | 500 |
| a. Free Acid Recovery | | | |
| Capital Cost | 42,600 | 52,800 | 79,200 |
| Operating Cost | 22,600 | 26,250 | 36,200 |
| Value of Nitric Acid Recovered | 10,900 | 19,600 | 49,000 |
| b. Recovery of Both Nitrate and Free Acid | | | |
| Capital Cost | 73,600 | 96,700 | 155,400 |
| Operating Cost | 36,100 | 51,300 | 95,700 |
| Value of Nitric Acid Recovered | 23,100 | 46,300 | 110,000 |

# APPENDIX

## COSTS OF INSTALLED PROCESS EQUIPMENT, CHEMICALS AND UTILITIES

Data derived in this study for the capital cost of the installed process equipment and the annual operating costs, excluding capital and labour charges, may be compared with equivalent data from Amdel (1966, 1970). Some differences are noted and explained but, in general, the comparison confirms the reliability of the estimates in this paper.

The costs of the installed process equipment for the standard flowsheet are detailed in Table A1 on the same cost basis as in the text. These costs are summarised in Table A2 to give the total cost of the installed process equipment. Estimates are also included in Tables A1 and A2 for the installation of essential utilities, though for an integrated fuel manufacturing plant the cost of some of these would be shared with the rest of the plant. Shared utilities are listed separately. Buildings, laboratory equipment and waste processing facilities are not included, nor are design and construction charges. The total costs of the installed process equipment and utilities for the 100, 200 and 500 tonnes/yr plants are respectively $A199,000, $A240,000, $A294,000.

A scale factor x can be calculated from the costs presented for the three sizes of plant considered, according to the formula:

$$\frac{C_1}{C_2} = \left(\frac{P_1}{P_2}\right)^x ,$$

where $C_1$ and $C_2$ are the costs of equipment and $P_1$ and $P_2$ are the capacities of the equipment for two sizes under consideration. The scale factor obtained is 0.25 for each interval, which may be compared with the generally accepted factor of 0.67 for chemical plant equipment which has been used by Amdel (1967, 1970). The factor is lower owing to the relatively small size of the plants considered and the cost of equipment is relatively insensitive to capacity in this range of sizes.

Amdel (1970) recently presented costs for a 500 tons $UO_2$/yr plant based on 8,000 hr/yr operation (7,200 in this report). Rotary kiln furnaces were selected for calcination, reduction and stabilisation, and the total cost of process equipment, excluding electrical, piping, instrument and installation costs, was estimated to be $A237,890. Comparative costs as estimated by Amdel, and in this report, are shown in Table A3 and there is good agreement between the Amdel total estimate and the figure of $A244,750 derived in this report.

The principal differences in costs are in Amdel's greater estimates for drying, calcination and reduction rotary kilns and smaller estimates for solvent extraction ancillaries and filtration. Our costs for rotary kiln furnaces are

## APPENDIX (continued)

based on examination of detailed quotations from the manufacturer. Amdel assumed very small tankage sizes for solvent extraction and assumed that the raffinate would be processed for nitric acid recovery, whereas our costs are based on large holding and neutralising tanks for raffinate and large interstage tanks. Also, the size of the rotary drum vacuum filter in the Amdel (1970) estimate is only 70 per cent of ours and filtrate storage and disposal equipment was not included in their estimate.

The costs of process chemicals and utilities involved in operating the plant with the standard flowsheet for throughputs of 100, 200 and 500 tonnes U/yr are detailed in Table A4. For a 500 tonnes/yr plant, we estimate the annual cost to be $A195,300, whereas Amdel (1970) obtained $A245,820. The main difference in chemical and utilities costs between the Amdel estimates and ours is in the nitric acid costs, where Amdel used $A222/ton for 69 per cent acid in drums and we used $A109/ton for 60 per cent acid in bulk. Nitric acid accounts for approximately two-thirds of the chemicals and utilities costs in our estimates and the saving involved in changing to bulk acid at lower concentration is therefore very important.

# TABLE A1

## CAPITAL COSTS OF INSTALLED PROCESS EQUIPMENT - STANDARD FLOWSHEET

| Item | 100 Tonnes/Yr | | 200 Tonnes/Yr | | 500 Tonnes/Yr | |
|---|---|---|---|---|---|---|
| | Unit Size | Cost, $A | Unit Size | Cost, $A | Unit Size | Cost, $A |
| 1. Yellow Cake Handling | | | | | | |
| Roller Conveyor, 20 in width, 4 in centres (1,500 lb capacity) | 15 ft | 150 | 15 ft | 150 | 15 ft | 150 |
| Tipping Box and Equipment | 144 $ft^3$ | 750 | 144 $ft^3$ | 750 | 144 $ft^3$ | 750 |
| | | 900 | | 900 | | 900 |
| 2. Dissolution - Batch | | | | | | |
| Tanks, Heated and Agitated, 2 off | 200 gal | 3,000 | 200 gal | 3,000 | 500 gal | 5,000 |
| Level Measurement | | 250 | | 250 | | 250 |
| Temperature Measurement and Control | | 1,450 | | 1,450 | | 1,450 |
| Nitric Acid Tank | 100 gal | 350 | 100 gal | 350 | 250 gal | 550 |
| Nitric Acid Pump | 10 gal/min | 450 | 10 gal/min | 450 | 25 gal/min | 600 |
| Nitric Acid Level Measurement and Pump Cut-Out | | 300 | | 300 | | 300 |
| Heat Exchanger (Simple Pipe Cooling Jacket) | 1 $ft^2$ | 200 | 1 $ft^2$ | 200 | 2.5 $ft^2$ | 250 |
| | | 6,000 | | 6,000 | | 8,400 |
| 3. Solvent Extraction - Mixer-Settlers | | | | | | |
| Construction, Framework and Assembly | | 10,000 | | 12,000 | | 16,000 |
| Agitators and Paddles | | 4,700 | | 5,000 | | 5,400 |
| Control Equipment and Panel | | 4,400 | | 4,400 | | 4,500 |
| Tankage - Strip | 500 gal | 800 | 1,000 gal | 1,400 | 3,000 gal | 3,000 |
| Scrub | 150 gal | 450 | 300 gal | 600 | 750 gal | 1,000 |
| Raffinate, 3 off, agitated | 200 gal | 2,250 | 350 gal | 3,000 | 1,000 gal | 5,400 |
| Product | 500 gal | 800 | 1,000 gal | 1,400 | 3,000 gal | 3,000 |
| Carbonate Wash | 150 gal | 450 | 300 gal | 600 | 750 gal | 1,000 |
| Solvent | 500 gal | 800 | 1,000 gal | 1,400 | 3,000 gal | 3,000 |
| Make-up, agitated | 250 gal | 850 | 500 gal | 1,200 | 1,500 gal | 2,200 |
| Pumps - S.X. to Precipitation, 2 off | 1 gal/min | 200 | 1 gal/min | 200 | 1 gal/min | 200 |
| Solvent Circulation, 2 off | 1 gal/min | 200 | 2 gal/min | 400 | 4 gal/min | 400 |
| Raffinate Discharge, 1 off | 6 gal/min | 200 | 12 gal/min | 250 | 35 gal/min | 300 |
| Make-up to Feed, 2 off | 15 gal/min | 700 | 35 gal/min | 1,100 | 100 gal/min | 1,400 |
| | | 26,800 | | 32,950 | | 46,800 |

continued...

TABLE A1 (continued)

| Item | 100 Tonnes/Yr | | 200 Tonnes/Yr | | 500 Tonnes/Yr | |
|---|---|---|---|---|---|---|
| | Unit Size | Cost, $A | Unit Size | Cost, $A | Unit Size | Cost, $A |
| 4. Precipitation - Single-Stage | | | | | | |
| Precipitation Vessel (agitated) | 12 gal | 300 | 25 gal | 350 | 65 gal | 500 |
| Control Equipment (pH, Temperature Recording) | | 2,000 | | 2,000 | | 2,000 |
| | | 2,300 | | 2,350 | | 2,500 |
| 5. Filtration - Rotary Drum | | | | | | |
| Vacuum Filter | | | | | | |
| Delay Tank (agitated) | 200 gal | 750 | 350 gal | 1,000 | 1,000 gal | 1,800 |
| Mono Pump | 50 gal/hr | 300 | 100 gal/hr | 300 | 250 gal/hr | 350 |
| Temperature Control | | 200 | | 200 | | 200 |
| Filter-Unit | 12 ft$^2$ | 13,500 | 25 ft$^2$ | 20,000 | 54 ft$^2$ | 24,000 |
| Filtrate Tanks, 2 off | 400 gal | 1,500 | 1,000 gal | 2,800 | 2,000 gal | 4,450 |
| Filtrate Discharge Pump | 20 gal/min | 500 | 50 gal/min | 600 | 100 gal/min | 700 |
| | | 16,750 | | 24,900 | | 31,500 |
| 6. Drying | | | | | | |
| Dryer Unit (incl. Roller Extruder) | 10 ft$^2$ | 20,000 | 20 ft$^2$ | 21,500 | 50 ft$^2$ | 26,000 |
| Transfer Hoppers, 2 off | 3 ft$^3$ | 500 | 5 ft$^3$ | 600 | 12 ft$^3$ | 700 |
| | | 20,500 | | 22,100 | | 26,700 |
| 7. Granulation | | | | | | |
| Jackson-Crockett Granulators (35 kg/hr) | 1 No. | 1,300 | 2 No. | 2,600 | 3 No. | 3,900 |
| Glovebox | 40 ft$^3$ | 400 | 80 ft$^3$ | 500 | 120 ft$^3$ | 600 |
| Transfer Hoppers, 2 off | 3 ft$^3$ | 450 | 5 ft$^3$ | 450 | 12 ft$^3$ | 600 |
| | | 2,150 | | 3,550 | | 5,100 |

continued...

TABLE A1 (continued)

| Item | 100 Tonnes/Yr | | 200 Tonnes/Yr | | 500 Tonnes/Yr | |
|---|---|---|---|---|---|---|
| | Unit Size | Cost, $A | Unit Size | Cost, $A | Unit Size | Cost, $A |
| 8. Calcination-Reduction and Stabilisation - Continuous Pulsed Bed | | | | | | |
| Calcination-Reduction Unit - construction and assembly | 6 in dia | 9,000 | 8 in dia | 12,000 | 10 in dia | 13,500 |
| Furnace | | 8,000 | | 9,000 | | 11,000 |
| Stabilisation Unit - construction and assembly | 6 in dia | 8,000 | 8 in dia | 11,000 | 10 in dia | 12,500 |
| Controls and Panel | | 9,000 | | 9,000 | | 9,000 |
| Interstage Transfer Hoppers, 3 off | 3 $ft^3$ | 1,100 | 5 $ft^3$ | 1,350 | 12 $ft^3$ | 1,600 |
| Product Hoppers, 3 off | 3 $ft^3$ | 600 | 5 $ft^3$ | 750 | 12 $ft^3$ | 900 |
| | | 35,700 | | 43,100 | | 48,500 |
| 9. Specific Utilities | | | | | | |
| Fume Scrubbing - jet scrubber incl. tanks, pump and piping | 1,000 $ft^3$/min | 6,500 | 1,000 $ft^3$/min | 6,500 | 1,000 $ft^3$/min | 6,500 |
| Nitric Acid Storage, Head Tank and Pumps | 2,000 gal | 3,000 | 4,000 gal | 4,300 | 6,000 gal | 5,400 |
| | | 9,500 | | 10,800 | | 11,900 |
| 10. General Utilities | | | | | | |
| Ammonia Storage | | | | | 2.5 tons | 3,500 |
| Ammonia Cracking, Inert Gas Generation, $N_2$ and $H_2$ Compression and Storage Receivers (generator is a standard size unit) | | 12,400 | | 12,400 | | 12,400 |
| Demineralised Water - unit and tankage | 75 gal/hr | 1,800 | 150 gal/hr | 2,750 | 380 gal/hr | 5,750 |
| Electricity | 200 kVA | 7,750 | 200 kVA | 7,750 | 200 kVA | 7,750 |
| Steam | 250 lb/hr | 1,750 | 335 lb/hr | 3,000 | 550 lb/hr | 3,900 |
| Ventilation and Filtration from powder handling boxes | 3,000 $ft^3$/min | 6,600 | 3,000 $ft^3$/min | 6,600 | 3,000 $ft^3$/min | 6,600 |
| | | 30,300 | | 32,500 | | 39,900 |

## TABLE A2

## SUMMARY OF CAPITAL COST OF INSTALLED PROCESS EQUIPMENT ($A)

| | 100 Tonnes/Yr | 200 Tonnes/Yr | 500 Tonnes/Yr |
|---|---|---|---|
| Yellow cake handling | 900 | 900 | 900 |
| Dissolution | 6,000 | 6,000 | 8,400 |
| Solvent extraction | 26,800 | 32,950 | 46,800 |
| Precipitation | 2,300 | 2,350 | 2,500 |
| Filtration | 16,750 | 24,900 | 31,500 |
| Drying | 20,500 | 22,100 | 26,700 |
| Granulation | 2,150 | 3,550 | 5,100 |
| Calcination-reduction | 35,700 | 43,100 | 48,500 |
| Specific utilities | 9,500 | 10,800 | 11,900 |
| Total Process Equipment Costs | 120,600 | 146,650 | 182,300 |
| Installation (40% Process Equipment Costs) | 48,100 | 58,850 | 72,800 |
| General Utilities | 30,300 | 32,500 | 39,900 |
| Total Installed Equipment Costs | 199,000 | 240,000 | 294,000 |

## TABLE A3

## COMPARISON OF COST ESTIMATES BY AMDEL AND THIS REPORT

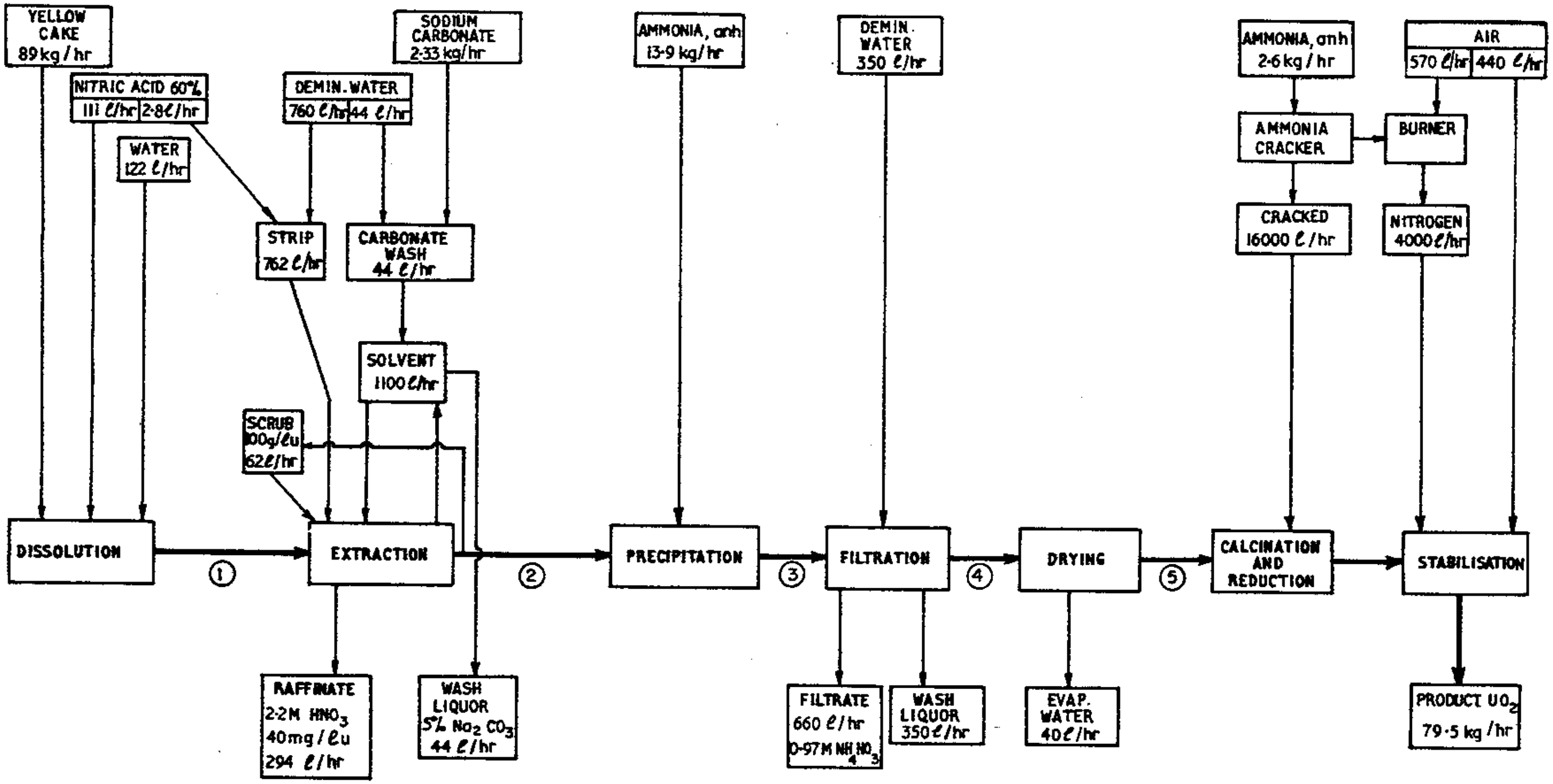
(Amdel 500 tons $UO_2$/yr; this report 500 tonnes U/yr)

| Equipment | Amdel | This Report |
|---|---|---|
| Dissolution | 8,650 | 9,300 |
| Solvent Extraction | 22,380 | 21,400 |
| Tanks and Pumps for Solvent Extraction | 3,750 | 25,400 |
| Precipitation | 1,710 | 2,650 |
| Filtration | 14,300 | 29,350 |
| Drying, Calcination and Reduction | 122,500 | 100,000 |
| Stabilisation | 25,500 | 25,000 |
| Services | 39,100 | 31,650 |
|  | $A237,890 | $A244,750 |

## TABLE A4

## ANNUAL COST OF PROCESS CHEMICALS AND UTILITIES - STANDARD FLOWSHEET

| Item | Unit Cost | 100 Tonnes/Yr |  | 200 Tonnes/Yr |  | 500 Tonnes/Yr |  |
|---|---|---|---|---|---|---|---|
|  |  | Annual Use | Cost, $A/Yr | Annual Use | Cost, $A/Yr | Annual Use | Cost, $A/Yr |
| Nitric Acid, 60% | $109/ton | 228 tons | 24,850 | 456 tons | 49,700 | 1,140 tons | 124,250 |
| Ammonia (anhydrous) | $160/ton | 23.4 tons | 3,750 | 46.8 tons | 7,500 | 117 tons | 18,700 |
| Tributyl Phosphate | $0.70/lb | 1,800 lb | 1,200 | 3,600 lb | 2,400 | 8,600 lb | 6,000 |
| Kerosene | $0.17/lb | 3,500 lb | 600 | 7,000 lb | 1,200 | 16,000 lb | 2,700 |
| Sodium Carbonate | $80/ton | 3.2 tons | 250 | 6.4 tons | 500 | 16 tons | 1,300 |
| Lime | $25.60/ton | 39 tons | 1,000 | 77.5 tons | 2,000 | 194 tons | 5,000 |
| Demineralised Water | $0.70/1,000 gal | $0.53 \times 10^6$ gal | 400 | $1.06 \times 10^6$ gal | 750 | $2.6 \times 10^6$ gal | 1,850 |
| Process Water | $0.20/1,000 gal | $3 \times 10^6$ gal | 600 | $6 \times 10^6$ gal | 1,200 | $15 \times 10^6$ gal | 3,000 |
| Fuel | $1.00/$10^6$ Btu | $1.3 \times 10^9$ Btu | 1,300 | $2.6 \times 10^9$ Btu | 2,600 | $4.8 \times 10^9$ Btu | 4,800 |
| Electricity | $0.025/kWh | 550,000 kWh | 13,700 | 700,000 kWh | 17,500 | $10^6$ kWh | 25,000 |
| Filter Cloth | $2.70/ft$^2$ | 220 ft$^2$ | 600 | 445 ft$^2$ | 1,200 | 1,000 ft$^2$ | 2,700 |
|  |  |  | 48,250 |  | 86,550 |  | 195,300 |



| TOTAL SUPPLIES REQUIRED | | | |
|---|---|---|---|
| YELLOW CAKE | 89 kg/hr | 2.14 tonnes/day | 640 tonnes/yr |
| NITRIC ACID, 60% | 114 ℓ/hr | 3.66 tons/day | 1,140 tons/yr |
| AMMONIA, anhydrous | 16.5 kg/hr | 0.39 tons/day | 117 tons/yr |
| SODIUM CARBONATE | 2.3 kg/hr | 0.053 tons/day | 16 tons/yr |
| WATER, DEMINERALISED | 1,278 ℓ/hr | 6,800 gal/day | $2.03 \times 10^6$ gal/yr |

| INTERSTAGE NO. | 1 | 2 | 3 | 4 | 5 |
|---|---|---|---|---|---|
| FLOWRATE, ℓ/hr | 233.5 | 700 | 700 | N.A. | N.A. |
| " , kg/hr ADU | – | – | 97.5 | 97.5 | 97.5 |
| U CONCENTRATION, g/ℓ | 300 | 100 | ~100 | 72% | 72% |
| M $NO_3^-$ | 3 | 0.97 | – | – | – |
| M $NH_4NO_3$ | – | – | 0.97 | 0.1 | 0.1 |
| WATER FLOWRATE, ℓ/hr | – | – | – | 40 | 1 |

FIGURE 1. STANDARD FLOWSHEET AND MASS BALANCE FOR A $UO_2$ PRODUCTION PLANT - CAPACITY 500 TONNES U/YR

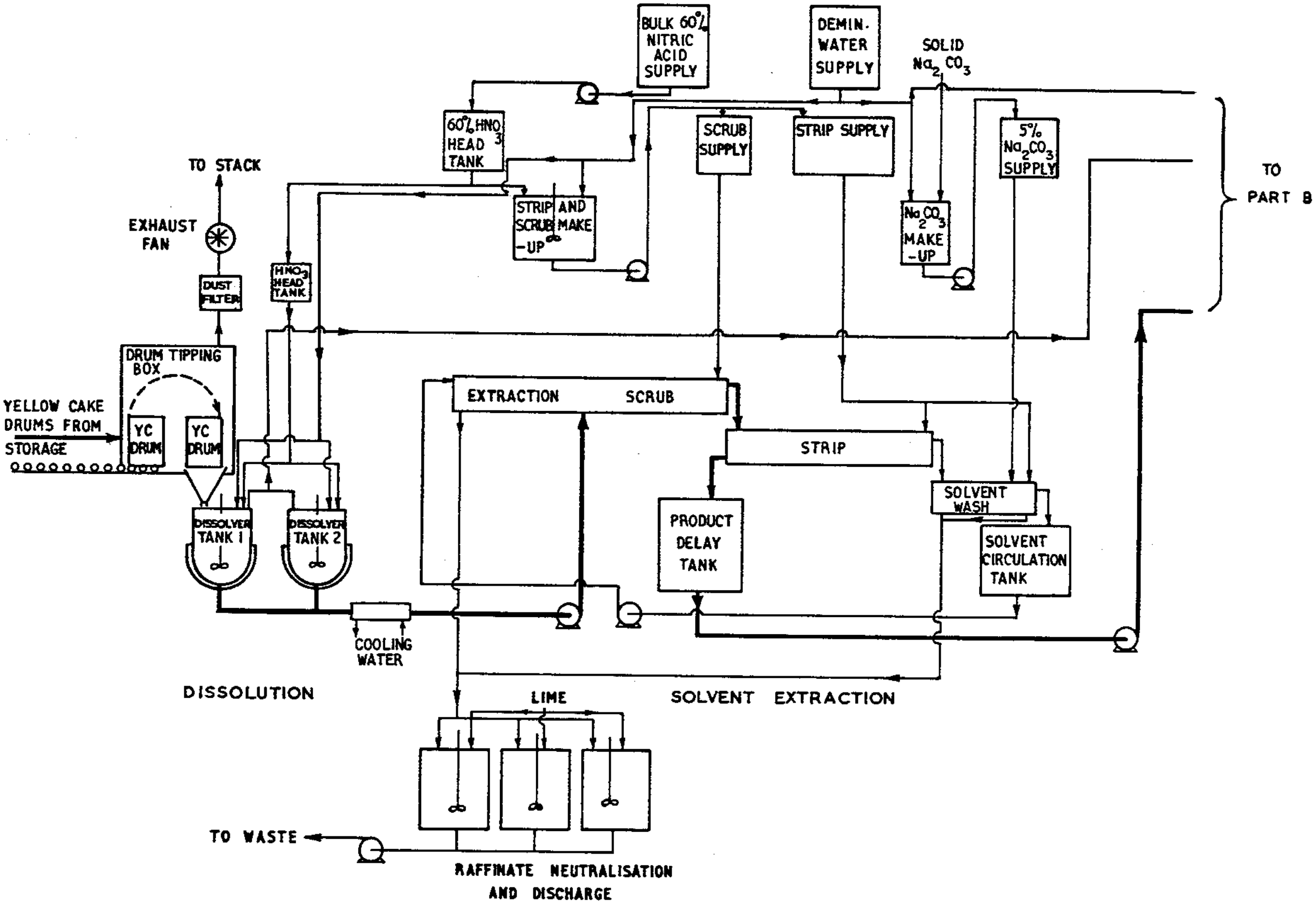


FIGURE 2. - PART A. - EQUIPMENT FLOWSHEET FOR A $UO_2$ PRODUCTION PLANT

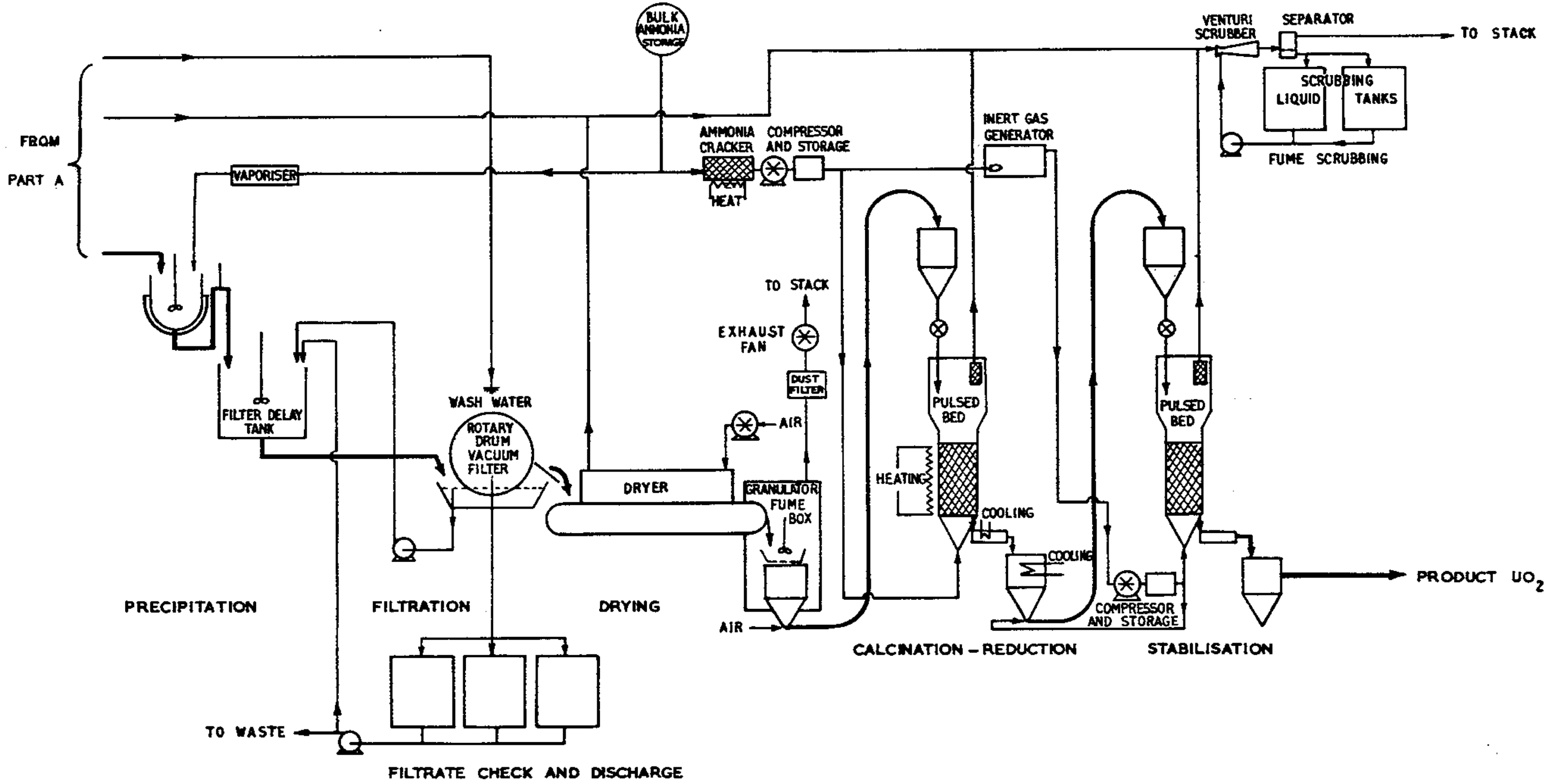


FIGURE 2. - PART B. - EQUIPMENT FLOWSHEET FOR A $UO_2$ PRODUCTION PLANT

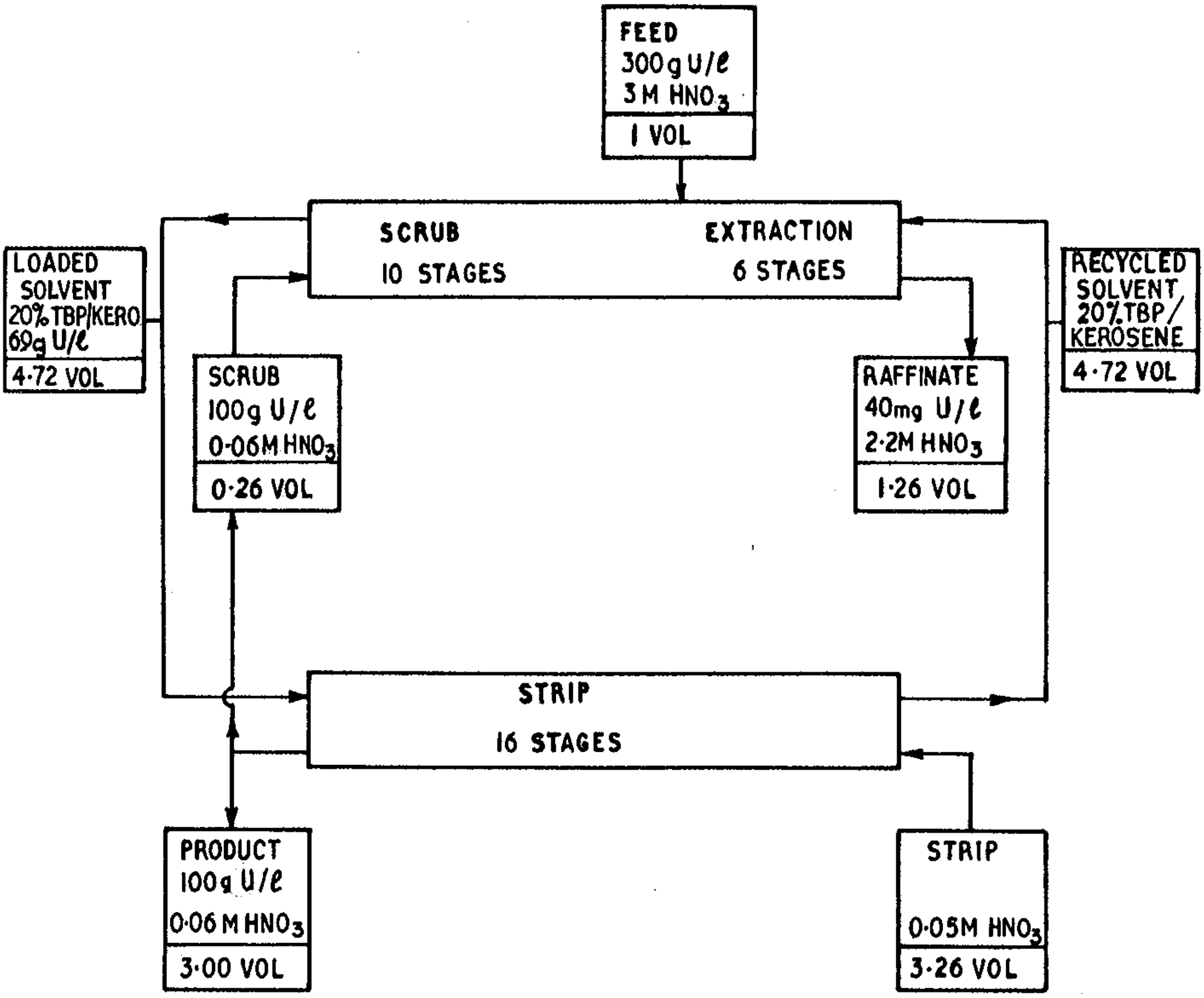


FIGURE 3. SOLVENT EXTRACTION FLOWSHEET FOR URANIUM PURIFICATION