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FEASIBILITY OF LARGE-SCALE NITROGEN-15
PRODUCTION FOR NUCLEAR REACTORS

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August 17, 1955

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FOR NUCLEAR REACTORS

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A B S T R A C T

Several processes are considered for the large-scale production of nitrogen-15 for (homogeneous) nuclear reactors.

It is concluded that one or several chemical exchange processes could be developed and a plant built to mass-produce 99% or 99.9% nitrogen-15 at less than \$5/gram, and possibly at close to \$1/gram, including initial costs amortized at 20% per year. The cost of chemicals, usually dominating in this type of process, can be eliminated as a controlling factor.

Nitrogen-15 is currently available in small quantities at about 70% assay for about \$300 per gram.

Note of July 1955:

This report was drafted in the early summer of 1954 and has not been brought up to date in wording or contents. Since its main conclusions remain valid, it is being issued unchanged to serve as a reference record of our past work. New processes, such as the NO-HNO₃ chemical exchange developed by Taylor, et al. at Columbia University and more recent versions of other chemical exchange reflux steps strengthen the confidence of the authors in the optimistic unit costs quoted in this report.

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FEASIBILITY OF LARGE-SCALE NITROGEN-15 PRODUCTION
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I. INTRODUCTION

The interest of nuclear scientists and engineers in low-cost nitrogen-15 was first mentioned to us in early 1954. R. A. Charpie of O.R.N.L. outlined the reasons for the interest and requested, on March 4, 1954, that we make a preliminary investigation of this isotope separation, specifically to determine whether the gaseous-diffusion process could yield N-15 at lower unit costs than those estimated for the chemical-exchange processes. Our conclusions were transmitted verbally to R. A. Charpie and others in May of 1954 and confirmed by letter of June 18, 1954 (KD-996). Pressure of production problems has prevented earlier issuance of this report which summarizes our study.

Current costs of N-15 in small quantities by chemical exchange were reported to be in the neighborhood of \$300/g., at an assay of around 70%. The commercial process is the same as used successfully by Urey and co-workers at Columbia University some years ago^(2,5). NH_4NO_3 and NaOH are purchased for refluxing, and NaNO_3 and NH_3 are discarded⁽¹⁸⁾. The equipment is the same or has been only slightly modified by the Eastman Company, and the process is stated to be quite satisfactory for present modest needs⁽¹¹⁾.

Estimates by the Materials Chemistry Division of O.R.N.L.⁽⁵⁵⁾ have indicated that the chemical costs could be decreased by resale of chemicals and by use of cheaper chemicals. From recent discussions with G. E. Clewett and J. S. Drury, it is established that the Materials Chemistry Division of O.R.N.L. is also envisioning chemical exchange processes of the "closed-reflux" type. Meetings have also been held with Dr. T. I. Taylor of Columbia University to encourage his investigation of the separation factor and an efficient reflux for the nitrogen oxide exchange process and several other promising ideas. It is the awareness of the combined interest and efforts of these experienced groups which makes us feel quite confident of our prediction of low-cost nitrogen-15.

This department has completed the requested preliminary process and economic survey for the large-scale separation of N-15. It is the purpose of this memorandum to record and discuss the results of the study which extended part time through most of March and part of April of 1954. It is hoped that the information can be combined with the results of experimental work and nuclear design evaluation to determine profitable courses of further planning and development.

II. SUMMARY AND CONCLUSIONS

Because of its low neutron capture cross section and high solubility of most of its nitrates, nitrogen-15 has interesting potential applications in the nuclear field. The current "market price" of about \$300 per gram has rendered its use uneconomical. However, this price is based upon present day needs calling for small product rates; considerable economy may be expected should large quantities be in demand. This study presents a brief survey of possible separation systems and order-of-magnitude evaluations of the more interesting processes. Although the major fields of interest have been examined, the study is by no means exhaustive. Other systems, as well as other modifications of the systems examined, can be conceived and may be more economical than those chosen. Much development work is necessary before any of the processes could be chosen and a plant constructed. Nevertheless, unit costs of 99 per cent nitrogen-15 on the order of \$1.00 to \$2.50 per gram seem attainable.

The task of aiming a preliminary evaluation of new processes at estimating eventual costs of mass production, after development, is obviously not an easy one. In the field of isotope separation, experience has shown it to be particularly hazardous to predict future developments and costs. Nevertheless, the authors have decided to conform with the request, since, for planning purposes, even a meagerly supported guess is still preferable to a complete lack of information. We feel that our conclusions can serve as justification for further development; they are not intended to be used for actual plant construction.

The processes studied were the gaseous diffusion process (as specifically requested by R. A. Charpie), a group of gas-liquid chemical exchange processes, ion exchange, and the gas centrifuge system. Of these, the chemical exchange processes appeared most promising so that the major effort was expended in this direction. A product assay level of 99 mole per cent nitrogen-15 and an eventual product rate of 50 metric tons per year were selected as goals. Relative costs of other assay levels and other production rates have been estimated and are shown in Figures 6 and 7.

Only four typical chemical exchange systems were examined, although many others are possible. Those chosen are described in Figures 2 - 5 and Tables I - X as well as in the discussion of this report. The gaseous diffusion process is described by Figure 8 and by Tables XI and XII. Ion-exchange and gaseous-centrifuge processes are described briefly in the discussion.

The chemical-exchange processes evaluated are as follows:

- 1) Closed-Reflux chemical exchange between NH_4NO_3 and NH_3 .
- 2) Chemical exchange between $(\text{NH}_4)_2\text{SO}_4$ and NH_3 .
- 3) Chemical exchange between NH_4OH and NH_3 .
- 4) Closed-Reflux chemical exchange between NO and N_2O_3 .

Figures 1 - 5 show schematically the above chemical exchange systems and also the "classical" system of Urey-Eastman.

All the ammonium-salt-ammonia exchange systems can be described in the following way. A solution containing ammonia and ammonium salt is passed countercurrent to ammonia gas in a suitable contacting device. At the product end of the plant, the ammonium salt is converted to ammonia, and the combined gases are returned to the cascade. At the bottom end of the plant, the ammonia is converted to the ammonium salt and this salt is returned to the cascade.

The "classical" system uses expensive chemicals for its reflux. The Eastman Company produces nitrogen-15 in small quantities by this process and current costs are stated to be in the neighborhood of \$300/g. at an assay of around 70%. In this report, the ammonia exchange processes selected were attempts to modify this process either by using cheaper chemicals, as in the cases of sulfate exchange and of hydroxide exchange, or to regenerate the chemicals by auxiliary steps and close the refluxes, as in the closed-reflux nitrate exchange.

These processes are all gas-liquid chemical exchange processes and utilize conventional gas-liquid contacting towers for the separation process. Without the benefit of detailed optimizations, it has been assumed to be most practical and economical to consider each separation process to operate close to atmospheric pressure and temperature conditions (about 10 psia. and 70°F.), except for the $\text{NO-N}_2\text{O}_3$ exchange (about 6 atms. and 32°F.).

The Urey-Eastman exchange takes place at 50 - 400 mm. Hg in order to avoid, as much as possible, the solution of ammonia in the liquid phase which decreases the effective separation factor. It is hypothesized⁽¹⁷⁾ that this reduction of separation factor is due to the exchange of ammonia gas with dissolved NH_3 rather than with NH_4^+ -ions. Operation in this low pressure range, of course, leads to low vapor densities and therefore large equipment for handling vapor mass flow. Furthermore, the refluxing of dissolved ammonia is usually much less expensive than refluxing of the ammonium ion. It is thus felt that an economic advantage has not been sufficiently established to justify the added complexities and costs of low-pressure operation. More detailed design and experimental studies should allow optimization of this variable. It is felt that even higher pressures than those assumed in this study may prove desirable.

A cost comparison of the chemical exchange processes is presented in Tables VII - IX. The estimates show a range of \$1.00 to \$2.50 per gram of nitrogen. This range may be compared to the costs of other processes - such as \$4.60 per gram by gaseous diffusion, \$4 per gram for chemicals alone by open-reflux ion exchange, and \$2,000 per gram by gaseous centrifugation. Thus, the economic advantage of the chemical exchange systems is obvious. More than this, however, may be drawn from the cost summaries. Open-reflux systems, such as the sulfate system, have relatively low initial costs but large operating costs whereas closed reflux systems, such as the nitrate system, draw their economic advantage from low operating costs. It may thus be concluded that the choice between these two types would tend to favor an open reflux for very short run operation

but a closed reflux for longer run operation. An examination of the cost breakdowns in Tables VIII and IX shows that the hydroxide system suffers most from large equipment and large electric power costs. These are items which could be reduced by an order of magnitude if the separation factor of that particular system is not too adversely affected by higher pressures. It may be seen that investigation of this one factor could either eliminate the hydroxide system or make it one of the major contenders. Likewise, the practicability of the nitrogen oxides system will depend upon the basis of its assumed separation factor and its extremely costly reflux. The uncertainties of the processes can affect not only the absolute magnitude of the final cost figure but the relative position of the possible systems as well.

The uncertainties just mentioned are listed along with the advantages and disadvantages of the primary schemes in Table X. It can be noted that the prime uncertainties of all the processes are concerned with the effect of pressure, temperature, and solution composition upon the separation factor and with the degree of completion of the reflux reactions. These points should comprise the initial effort of any development program. After these factors are better known, an elimination of processes can be made more confidently, thus allowing the effort to be concentrated on those having the most promise. It should not be forgotten, however, that other chemical exchange systems are possible which should be similarly checked.

Some of the alternatives possible are mentioned briefly in the discussion. For example, the refluxes of the nitrate systems might be modified so that cheaper chemicals are purchased rather than closing the refluxes. On the other hand, other schemes than those adopted for closing the reflux might be used. Many anions other than those chosen are, of course, possible, e.g., a closed reflux scheme should be possible with either the carbonate or the chloride. Thus, the scope of this report is a brief survey of representative processes and is not intended to be definitive.

The designs and cost estimates, it need hardly be emphasized, are supported only to a very limited extent by experimental data. No attempt has therefore been made to perform elegant optimizations of design variables, such as operating temperatures, pressures, feed rates, etc. It was decided from the start that no single process could be spotlighted as the economical process in the light of today's knowledge. It was hoped, rather, that some individual processes might be eliminated as too costly or involved, that a general cost range might be established for product from one of several processes, and that from such a study, certain specific needs for development might become apparent.

III. DISCUSSION

There are many methods that could be used for the separation of nitrogen-15. The accompanying bibliography contains references for the following separation processes: chemical exchange (both gas-liquid and gas-gas)⁽¹⁻²⁵⁾; gaseous diffusion, mass diffusion, and sweep diffusion⁽²⁶⁻³⁰⁾; thermal diffusion⁽³¹⁻³⁴⁾; ion exchange⁽³⁵⁻³⁸⁾; and electrolysis⁽³⁹⁾.

Seven separation systems have been evaluated in this report, these being four chemical exchange processes, one gaseous diffusion process, one ion exchange process, and one gaseous centrifuge process. The emphasis on the chemical exchange processes reflects the authors' opinion that they will prove to be the most economical. The other processes have been evaluated in less detail and are included for the sake of comparison.

A. Chemical Exchange Methods

It has been demonstrated by several workers and by actual small-scale plant experience^(18,20) that nitrogen, considerably enriched in the nitrogen-15 isotope, can be produced by countercurrent chemical exchange methods. Although any compound of nitrogen is potentially a candidate for chemical exchange reactions, ammonia and its compounds have received the bulk of attention.

The exchange reaction of the general type

where the anion is unspecified, was first proposed by Urey and Grieff⁽¹⁾ in 1935. Although at that time, doubt was expressed about its feasibility, Urey and his co-workers^(2-5,7), Thode⁽¹⁴⁾, and Kirshenbaum⁽¹⁷⁾, in this country, Clusius and co-workers^(12,13) and Becker and Baumgartel⁽¹⁵⁾ in Germany, and Sugimoto, et al.⁽²⁰⁾ in Japan have worked on this exchange reaction since the original proposal. The anions usually have been the sulfate, nitrate, or hydroxide. The solvent used has been water, except for a few tests in which it was ethyl alcohol.

In addition to the ammonium salt-ammonia exchange systems, various other compounds of nitrogen have been examined for possible production of N-15 enriched nitrogen. Leifer⁽²¹⁾, Ogg, et al.^(22,24), Taylor and Spindel⁽²³⁾, and Taylor, et al.⁽²⁵⁾ have worked on chemical exchange using the oxides of nitrogen. Nisina, et al.⁽⁸⁾, Norris, et al.⁽⁹⁾, and Joris⁽¹⁰⁾ investigated exchange reactions between gaseous nitrogen and various nitrogen containing liquids. Roberts, Thode, and Urey⁽⁶⁾ and Becker and Vogell⁽¹⁹⁾ studied the exchange reaction between HCN and cyanides.

1. General Discussion

Of the four chemical exchange systems evaluated in this report, three are ammonia exchange systems and the other a nitrogen oxide

exchange system. As was mentioned before, the bulk of the chemical exchange work has been done with the compounds of ammonia. In contrast, the evaluation of the nitrogen oxides exchange system is based upon very meager data. Hence, the following general discussion will deal with ammonia exchange systems.

For the general exchange reaction between ammonia and an ammonium salt, five anions (nitrate, sulfate, chloride, hydroxide, and carbonate) have been considered in aqueous solutions; and one anion (hydroxide) has been investigated in an alcohol medium. The non-aqueous solvent appeared to give no advantage over water and can be dropped from further consideration because of the cost of the required inventory.

The high solubility of ammonia in water prevents the exchange reaction from proceeding as originally written at any finite pressure. Instead, two competing reactions are taking place at once, for which the equations may be written as

where $\text{NH}_3 (\text{aq})$ represents all the gaseous ammonia dissolved in water without specifying its form. For both of the reactions, the equilibrium constant, k , is greater than one, thus favoring the concentration of N-15 in the aqueous phase.

The separation factor for the combination of reactions I and II depends upon the equilibrium constants of the individual reactions and upon the relative amounts of nitrogen exchanging by each reaction. The amount of nitrogen exchanged by each reaction will depend upon the molar concentration of ammonium salt and of ammonia gas. The ultimate solubility of both the salt and the gas depend upon the partial pressure of ammonia in the gas phase.

By extrapolation to zero pressure, Kirshenbaum, et al.⁽¹⁷⁾ found the separation factor for reaction I in 60 per cent nitrate solution to be 1.034. The separation factor for reaction II, in the total absence of ammonium salt, has been placed at 1.005 - 1.006 by Urey and Aten⁽²⁾. Separation factors for the combined reactions have been determined by other investigators to be such values as 1.022 for nearly saturated sulfate solution⁽⁴⁾, 1.027 for nearly saturated chloride solution⁽²⁾, and 1.021 for nearly saturated nitrate solutions⁽¹⁴⁾, all at low ammonia partial pressures.

In addition to the rather favorable equilibrium relations, there are other reasons favoring the extensive development of ammonia-ammonium salt exchange systems. Ammonia is abundant and relatively

inexpensive. The cascade sections do not have to be protected from inleakage of small amounts of air or water. The wide variety of compounds that ammonia forms with acids allows a large variation in possible exchange systems and reflux reactions. Finally, there is a great deal of experience in handling ammonia and its many reaction products.

All ammonium salt - ammonia exchange systems have the same general construction. The complete process may be described in the following way. A solution containing ammonia and ammonium salt is passed countercurrent to ammonia gas in a suitable contacting device. At the product end of the plant, the ammonium salt is converted to ammonia, and the combined gases are returned to the cascade. At the bottom of the plant, the ammonia is converted to the ammonium salt and this salt is returned to the cascade. On an overall basis, the reflux reactions can be considered as the acceptance of an anion by ammonia from a suitable donor and later the rejection of the anion at the other end of the plant to a suitable acceptor. Thus, the process consists of the transfer of a given anion from one compound to another through an intermediate step.

2. "Classical" System

The chemical exchange system as proposed by Urey⁽⁴⁾ was that between NH_4NO_3 and NH_3 with the reflux reaction as follows:

A modified version of this scheme was used by several workers^(13,18,20) for the production of nitrogen-15 enriched nitrogen. The modification consists of eliminating the waste-end reflux by withdrawing and discarding NH_3 and adding fresh NH_4NO_3 . This modified system is shown schematically in Figure 1.

3. Description of Systems Evaluated

The chemical exchange systems evaluated in this report are:

- a) Closed Reflux Chemical Exchange Between NH_4NO_3 and NH_3 . This system retains the exchange of the classical system but attempts to avoid prohibitive costs of the large chemical reflux streams by closing the refluxes. The closed-reflux system as shown in Figure 2 utilizes $\text{Ca}(\text{OH})_2$ at the product-end reflux to convert the NH_4NO_3 to NH_3 as follows:

This reaction is also used in Solvay process plants for the recovery of NH_3 . The excess $\text{Ca}(\text{OH})_2$ is filtered and recycled and the $\text{Ca}(\text{NO}_3)_2$ solution is concentrated and forwarded to the waste-end reflux.

The waste-end reflux combines the $\text{Ca}(\text{NO}_3)_2$, received from the product-end reflux, with CO_2 to convert the NH_3 to the NH_4NO_3 phase as follows:

This conversion takes place in two steps. First, the CO_2 and NH_3 are absorbed in H_2O , forming $(\text{NH}_4)_2\text{CO}_3$. The $(\text{NH}_4)_2\text{CO}_3$ is then reacted with $\text{Ca}(\text{NO}_3)_2$ to produce NH_4NO_3 and CaCO_3 . The CaCO_3 is filtered and decomposed in rotary kilns to produce CaO and CO_2 . The CaO is reacted with water to form $\text{Ca}(\text{OH})_2$ which is sent to the product-end reflux while the CO_2 is recycled to the absorption tower. The NH_4NO_3 solution must be concentrated before being returned to the cascade. The NH_3 obtained from the evaporation step is recycled to the absorption tower.

An advantage in using NH_4NO_3 as the ammonium salt is that its solubility is greater than that of other ammonium salts. Also, an increase in the partial pressure of ammonia does not repress the solubility of ammonium nitrate as much as it does that of other ammonium salts. This means that for any given temperature and pressure, the NH_4NO_3 system will have a higher separation factor and therefore a smaller cascade for a specified product rate than most other ammonia systems.

- b) Chemical Exchange Between $(\text{NH}_4)_2\text{SO}_4$ and NH_3 . This system may be regarded as intermediate between the classical and the closed reflux nitrate systems. The high chemical costs of the classical system are reduced by the use of cheaper chemicals while the complexity of the closed reflux with its attendant investment cost is avoided.

The $(\text{NH}_4)_2\text{SO}_4$ - NH_3 system as shown in Figure 3 has essentially the same product-end reflux as the closed reflux nitrate process except that CaSO_4 is produced instead of $\text{Ca}(\text{NO}_3)_2$ as follows:

The CaSO_4 is considered to have no value and is discarded; however, $\text{Ca}(\text{OH})_2$ slurry has taken the place of the more expensive NaOH of the classical system.

The waste-end reflux is the neutralization of NH_3 by H_2SO_4 as follows:

which, if compared with the Eastman system, again shows the replacement of an expensive chemical, NH_4NO_3 , by a cheap chemical, H_2SO_4 .

The exchange section of the sulfate system will be larger than the corresponding section of the nitrate system because the lower solubility of the ammonium sulfate results in a lower separation factor.

- c) Chemical Exchange Between NH_4OH and NH_3 . The refluxes of this system are relatively simple involving neither the complexity of a closed reflux system nor the necessity of purchasing reflux chemicals. On the other hand, due to a lower separation factor (about one-third that of either the nitrate or the sulfate system), the exchange section is much larger than for the other systems.

The NH_4OH - NH_3 system as shown in Figure 4 utilizes the distillation of NH_3 from solution for the product-end reflux as follows:

The waste-end reflux is the absorption of NH_3 by water in a short length of the towers.

One advantage of this system is that mild steel is suitable as a construction material.

- d) Closed Reflux Chemical Exchange Between NO and N_2O_3 . The nitrogen-oxides exchange system, illustrated in Figure 5, is different in a number of ways from the ammonia systems. It consists of exchange between NO (gas) and N_2O_3 (liquid) at a higher pressure (6 atm.) and a lower temperature (ca. 32°F .) than the ammonia-ammonium salt exchange pressures. Due to its higher separation factor ($\alpha-1 = 0.050$) as well as to the high pressure, the exchange system itself is small by comparison with the NH_3 systems. The waste-end reflux is quite easy, being the oxidation of NO to

NO_2 and liquefaction forming N_2O_3 ($\text{NO}_2 + \text{NO}$). This reaction takes place rapidly and spontaneously, even in air, although the use of pure oxygen has been assumed to prevent excessive losses of NO_2 with the inert gases. The product-end reflux, on the other hand, is complex and may not even be practicable. The problem is to reduce N_2O_3 (or NO_2) to NO without the formation of N_2 . It has been assumed that this reduction is possible by reaction of the NO_2 with FeSO_4 and H_2SO_4 forming NO , $\text{Fe}_2(\text{SO}_4)_3$ and H_2O . Further, the assumption has been made that a cell can be devised to regenerate the ferrous sulfate electrolytically from the ferric sulfate.

4. Bases for Evaluation

A survey was made of the available data published on the chemical exchange processes evaluated in order to provide firm bases for design. The bases selected are as follows:

- a) For the sulfate and nitrate systems, the relation between separation factor and aqueous phase composition proposed by Kirshenbaum⁽¹⁷⁾ was used. For the ammonia-water exchange system, the separation factor was taken from the work of Urey and Aten⁽²⁾. For the nitrogen oxides system, the separation factor was estimated by Taylor⁽⁴¹⁾.
- b) The temperature for the ammonia systems was arbitrarily selected as the approximate temperature at which the separation factors were determined. For the oxide system, the temperature was selected to be above the critical temperature of NO and below the normal boiling temperature for N_2O_3 .
- c) The pressure selected for the ammonia exchange systems was a compromise between a low pressure (high separation factor), a high pressure (low compression and cooling costs), and a desire not to extrapolate too far beyond the pressures used in the separation factor determinations. The latter consideration carried the most weight in the final decision. The pressure selected for the oxide exchange system was an arbitrarily selected value that appeared reasonable and practicable.
- d) The physical properties for the system $\text{NH}_3\text{-NH}_4\text{NO}_3\text{-H}_2\text{O}$ were taken primarily from the work of Worthington, Datin, and Schultz⁽⁴²⁾ and that of Hunt⁽⁴³⁾. The physical properties for the system $\text{NH}_3\text{-(NH}_4)_2\text{SO}_4\text{-H}_2\text{O}$ were taken from Hill and Loucks⁽⁴⁴⁾ and from Seidell⁽⁴⁵⁾. Since inadequate vapor pressure data were available, the vapor pressure was assumed to be equivalent to the pressure for the nitrate system.
- e) Sieve-plate columns are used for the gas-liquid contacting in the cascade for all the exchange processes evaluated in this report. Although sieve-plate columns have a relatively narrow range of

velocities for efficient operation, they are cheap, and the flow rates could be varied by the number of parallel columns in operation, rather than by the velocities through all the columns. The assumed stage efficiencies and flows were felt to be reasonable engineering estimates but somewhat on the optimistic side (see, for example, Perry⁽⁴⁶⁾, Jones and Pyle⁽⁴⁷⁾, and Arnold, Plank, and Schoenborn⁽⁴⁸⁾). The materials of construction selected were based on corrosion data in Perry⁽⁴⁹⁾ and Chemical Engineering⁽⁵⁰⁾.

- f) The thermodynamic properties of ammonia for use in compression and cooling calculations were taken from Perry⁽⁵¹⁾. The vapor-liquid properties for the system $\text{NH}_3\text{-H}_2\text{O}$ were taken from Hougen and Watson⁽⁵²⁾ and from Perry⁽⁵³⁾. These data could be used directly for the hydroxide system and were assumed to apply to the other systems. This assumption was checked at two points for the sulfate system from data in the International Critical Tables⁽⁵⁴⁾; however, at conditions far from the actual product-reflux-tower conditions.

5. Process Comparison

Tables I-X summarize and compare the four chemical exchange processes which have been calculated. Process bases, major plant equipment, operational requirements, costs, and advantages and disadvantages are compared in these tables.

Table I gives the process bases comparison. In this table, the product, feed, and waste rates are given, the reflux and supplementary reactions are summarized and the process and equipment variables are listed. It should be emphasized that few of the reactions discussed are quantitative, in practice or in theory, even though the reactions have been written in this manner. Factors such as necessary multi-step reactions, required excess of reagents, resultant losses of desired chemicals, or required equipment complexity, may make any of the reactions economically undesirable.

One of the most important design variables is the static pressure level of the tower in the ammonia exchange sections. As mentioned previously, as the pressure is increased, the separation factor decreases. A decrease in separation factor results in a cascade that is required to handle a greater total molar flow and in a somewhat larger reflux system. However, as the pressure level goes up, the towers can handle more flow per unit area and the energy requirement for gas-phase compression and cooling are greatly reduced. Final economic balances to select optimum tower pressures are beyond the present knowledge of the exchange systems. It is felt, though, that higher pressure levels than used in this study will be optimum.

The major plant equipment for the chemical exchange processes evaluated is listed in Tables II-V. The operational requirements are listed in Table VI. For the purpose of discussing the equipment,

a chemical exchange plant may be divided into four sections. These are: the exchange area, the product-end reflux reaction, the waste-end reflux reaction, and the cascade auxiliary sections. For the ammonia systems, which will be discussed together, there are many similarities between corresponding sections.

The exchange sections for the ammonia processes could consist of the following major items:

1. Exchange Towers - countercurrent gas-liquid contacting towers in which the reversible exchange of nitrogen-15 takes place.
2. Upflow Pumps - centrifugal pumps provided to move the aqueous phase between towers or from a blender to a tower.
3. Blenders - enclosed mixers for blending the aqueous phases from parallel towers before another row of parallel towers.
4. Blender Pumps - centrifugal pumps provided to move the aqueous phase from one tower to a blender.
5. Blowers - centrifugal compressors for transferring the gas phase from one tower to the next tower in the same line.
6. Coolers - finned-tubular gas coolers for maintaining isothermal conditions in the exchange cascade.
7. Feed System - including such items as unloading and storage facilities for liquid ammonia and heaters for the ammonia gas after it is expanded to tower pressure.
8. Minor Equipment - such items as instruments to control liquid levels, gas flows, temperatures, and other cascade variables; and piping for aqueous and gas phase process lines as well as instruments, cooling, and other auxiliary lines.

The top-reflux sections for the ammonia processes could consist of the following items:

1. Towers - distillation towers for the separation of ammonia from water or from aqueous salt solutions.
2. Tower Auxiliaries - such items as condenser, reboiler, reflux pump, product and waste pumps, feed preheater and other distillation tower auxiliary equipment.
3. Reflux Return System - the blowers and coolers or evaporators which are needed to return the tower top-product to the cascade as reflux.

4. Mixers - liquid mixers for mixing the cascade aqueous flow with the lime slurry for refluxing of the nitrate or sulfate system.
5. Product System - facilities for withdrawing the product from the cascade and converting the product to the desired form.
6. Minor Equipment - such items as piping, instruments, and minor auxiliaries necessary for the proper functioning of the other units.

The bottom-reflux systems require different equipment for each process. In view of this, it is best to list each process separately. For the nitrate system the major items for the waste-end reflux are as follows:

1. Absorbers - gas liquid contacting devices used to form an ammoniated solution of ammonium carbonate.
2. Reactors - agitated liquid reactors for reacting ammonium carbonate and calcium nitrate.
3. Crystallizers - used to aid crystal formation of the CaCO_3 .
4. Filters - continuous filters used for clarifying the effluent and washing and dewatering the crystals from the crystallizers.
5. Steam Jet Ejectors - used for operation of the filters.
6. Evaporators - used to concentrate the effluent from the filters.
7. Crystallizers - used to separate ammonium nitrate crystals from the concentrated effluent obtained from the evaporators.
8. Centrifuges - continuous centrifuges for recovering the crystals of ammonium nitrate.
9. Driers - low-temperature driers for drying the ammonium nitrate crystals from the centrifuges.
10. Stills - fractionating columns used to recover ammonia from the condensate from the steam-jet ejectors and evaporators.
11. Mixers - used to mix the dried ammonium nitrate crystals with a water-ammonia mixture to form the cascade upflow.
12. Ammonia Condensers - used to liquefy the gaseous ammonia downflow.

13. Cascade Waste System - facilities for handling, storing, and shipping ammonia from the cascade.
14. Minor Equipment - pumps, blowers, coolers, and heaters, storage and surge tanks, solid conveyors, dust recovery equipment, instruments, and piping.

For the sulfate system the following is the major equipment for the waste-end reflux:

1. Absorbers - gas-liquid contacting devices in which ammonia, diluted in air, is absorbed into sulfuric acid.
2. Evaporators - used to concentrate the liquid from the absorbers.
3. Cascade Waste System - including facilities for handling, storing, and shipping ammonium sulfate from the cascade.
4. Minor Equipment - including pumps, blowers, coolers, and heaters, storage and surge tanks, instruments and piping.

For the hydroxide system the following is the major equipment for the waste-end reflux:

1. Absorbers - gas-liquid contacting devices in which ammonia is absorbed by water.
2. Cascade Waste System - facilities for handling, storing and shipping ammonia from the cascade.
3. Minor Equipment - pumps, blowers, coolers, ammonia condensers, storage and surge tanks, instruments and piping.

The auxiliary system for each process includes such important items as refrigeration, electrical systems, steam plants, cooling water supplies, and storage areas. In addition, the nitrate system has certain necessary auxiliary equipment required for the closed nature of its reflux. However, the open reflux sulfate and the hydroxide systems have no additional auxiliary equipment.

For the nitrate, some of the additional auxiliary equipment that might be required is as follows:

1. Preheaters - used to dry and preheat the CaCO_3 sludge before calcining and to act as coolers for the gases from the calciner.
2. Burners - rotary or shaft type direct-fired lime-kilns used to calcine dried CaCO_3 sludge.

3. Coolers - rotary coolers used to cool the burned lime and to preheat the combustion air.
4. Hydrators - used to form milk of lime for use in the top-product reflux.
5. Blowers - used to move the gases from the preheaters to the CO_2 recovery system.
6. Coolers - used to cool the gases before the CO_2 recovery system.
7. Absorbers - countercurrent gas-liquid contacting devices used to recover CO_2 from the gases from the kilns.
8. Desorbers - countercurrent gas-liquid contacting devices in which the recovered CO_2 is separated from the absorbent.
9. Solvent Cleanup and Makeup System - including such equipment as is needed to prepare, store, and purify the absorbent. This system would also contain equipment for reducing the absorbent loss that is caused by saturation of the gas by the absorbent.
10. Evaporators - used to concentrate the aqueous solution of $\text{Ca}(\text{NO}_3)_2$ obtained from the top-reflux section for use in the bottom reflux section.

6. Cost Comparison

Table VII gives a cost summary of the chemical exchange processes while Table VIII and IX give breakdowns of the construction and operational costs, respectively. The cost summary shows that for a plant to produce 50,000 Kg. of nitrogen-15 per year an initial investment cost of 80 to 170 million dollars and a five year total operational cost of 150 to 400 million dollars would be required, depending on the process selected. This results in a unit cost of about 1 dollar per gram for the $\text{NH}_3\text{-NH}_4\text{NO}_3$ and $\text{NO-N}_2\text{O}_3$ exchanges and about 2 dollars per gram for the other processes.

The equilibrium time varies from 7 to 400 days. This variation is mainly due to the value of the separation factor which is low for the $\text{NH}_3\text{-NH}_4\text{OH}$ exchange, corresponding to a long equilibrium time, and high for the $\text{NO-N}_2\text{O}_3$ exchange, corresponding to a short equilibrium time. A cost penalty for the deferred production due to equilibrium time has been included.

As is to be expected, the $\text{NH}_3\text{-(NH}_4)_2\text{SO}_4$ process has a relatively low initial cost at the expense of a high operating cost due to the large purchases of even relatively cheap chemicals. On the other hand, although the $\text{NH}_3\text{-NH}_4\text{NO}_3$ process with its closed reflux is more complex, the expense of the additional equipment is much lower than the eliminated chemical costs. This would mean, from an economic viewpoint, that for short amortization periods an open-reflux system would be more preferable; while the closed reflux systems become more attractive as the amortization period is increased.

There are no chemical costs (outside of feed and water) for the $\text{NH}_3\text{-NH}_4\text{OH}$ system. However, the additional costs of equipment and power for handling the larger flows counterbalance the simplicity of the refluxes.

Practically the entire cost of the $\text{NO-N}_2\text{O}_3$ system is in the product-end reflux. Specifically, this is the investment and power cost of electrolytic cells. The high cost of this reflux, coupled with its previously mentioned uncertainty, makes the development of a simpler reflux very desirable.

The advantages, disadvantages, and uncertainties of the chemical exchange processes are listed in Table X. The cost advantages and disadvantages given depend to a large extent on the resolution of the uncertainties.

There has been some speculation that the separation factor of the $\text{NH}_3\text{-NH}_4\text{OH}$ exchange system may be independent of pressure. In this report, it has been assumed that the separation factor would decrease as the pressure increases for all the NH_3 exchange processes. Should experience prove this assumption to be invalid for the $\text{NH}_3\text{-NH}_4\text{OH}$ exchange, the system could be greatly reduced in size and cost by operation at high pressures. As was stated previously, the pressure chosen for the exchange sections, 10 psia., was close to the pressures at which the separation factors were determined.

Another uncertainty which could greatly influence the cost picture is the product-end reflux of the $\text{NO-N}_2\text{O}_3$ exchange system. The problem is to reduce N_2O_3 (or NO_2) to NO without the formation of any other nitrogen compounds or nitrogen. In this report, it has been assumed that this could be accomplished by the reaction of NO_2 with FeSO_4 and H_2SO_4 and that a cell could be devised to regenerate FeSO_4 from the $\text{Fe}_2(\text{SO}_4)_3$ formed. Not only is this reflux very uncertain, but its cost is essentially the whole cost of the system. It is therefore apparent that, although an extensive development program might be required to obtain a successful reflux, the pay off could be great.

A very important uncertainty which is shared by all the chemical exchange processes and which would affect costs greatly is the completeness of the product-end reflux. In the final sectionalized plant, there will be one product reflux plus several product-end refluxes at intermediate assays. The higher the assay level the more critical is the requirement that the reflux be complete. This is extremely important because the loss of highly enriched material means a decrease in production and may even make the desired product assay level unobtainable. An important consideration in the design of these top refluxes may be the decomposition of ammonium salts. Most of the ammonium salts are rather susceptible to thermal decomposition forming various decomposition products that might be lost from the cascade.

7. Alternate Product Rates and Assay Levels

In Figure 6, the variation of the unit cost of product with the product assay level is shown. An assay level of 99 per cent was arbitrarily selected as a base. For enrichments much greater than this selected level, there is only a slight increase in the unit cost; since the great part of the costs are incurred in the refluxes and in reaching this base assay from the feed. An additional increase in product assay requires only the addition of relatively small towers. However, the penalty for deferred production, not included in Figure 6, is increased at higher assays.

In Figure 7, the variation of the unit cost of production with product rate is shown. The savings possible through large scale operation are well known. Larger items and duplicated items allow savings to be made in construction of the plant. In operation, savings can be had in such items as labor charges, steam, and electricity as the required amounts of each increases. However, the initially selected product rate of 50 metric tons per year was high enough so that any further increases in production would not result in substantial savings. For the same reason, a large decrease in product rate would have to be made before the unit cost increased appreciably. From the figure, it is seen that the unit costs of product from the processes having the greater portion of costs in the reflux are less sensitive to the product rate.

It should be mentioned that both Figures 6 and 7 are probably more accurate than the basic unit cost numbers.

8. Alternate Chemical Exchange Systems

Although many compounds of nitrogen could be used for chemical exchange reaction, ammonia and its compounds have received the bulk of attention. The ammonia exchange systems evaluated in this report were not selected by any detailed process of elimination but rather represent modifications of the classical system. It is therefore quite possible that other schemes might be more promising. Some alternate or additional modifications that one might make are as follows:

a) Nitrate Exchange System with Inexpensive Chemicals

It might be possible to modify the classical system so that the sodium hydroxide and nitric acid used in the refluxes would be regenerated by treating the NaNO_3 formed in the bottom reflux.

The reactions involved would be as follows:

In essence, the process is modified so that the much cheaper chemicals, $\text{Ca}(\text{OH})_2$ and H_2SO_4 are substituted for the more expensive chemicals, NaOH and HNO_3 . However, the second reaction listed above is quite uncertain and would require special operating conditions. This scheme might be cheaper than the NH_3 -(NH_4)₂ SO_4 system evaluated which uses $\text{Ca}(\text{OH})_2$ and H_2SO_4 in the reflux reactions. It would depend on the benefit of the smaller nitrate cascade versus the disadvantage of the auxiliary chemical steps needed.

b) Alternate Closed Reflux Nitrate System with Sodium Salts

Instead of the closed refluxes used in the NH_3 - NH_4NO_3 system, the following closed refluxes might be used:

At the waste-end NH_3 could be converted to NH_4NO_3 as follows:

The NaHCO_3 is precipitated from the cold, concentrated solution and the NaOH for the product reflux is obtained as follows:

The NaOH is sent to the product-end reflux closing the circuit.

The CaCO_3 is filtered off and may be calcined and slaked to regenerate the $\text{Ca}(\text{OH})_2$ required in the above reaction as follows:

c) Closed-Reflux Carbonate Exchange System

Most of the regenerative or closed-reflux systems mentioned make use of the precipitation of calcium carbonate from a solution and the regeneration of lime from the precipitate. This suggests that the carbonate ion might be used directly as the exchange anion. This system was suggested by H. C. Urey⁽¹⁶⁾ who conjectured on the formation of a carbamate ion and its effect on the separation efficiency. The reflux reactions would then be as follows:

d) Closed Reflux Chloride Exchange Systems

The NH_3 - NH_4Cl exchange might be used, involving the following reflux steps:

The NaCl formed could be electrolyzed to recover NaOH, H_2 , and Cl_2 . The H_2 and Cl_2 would then be recombined to form HCl. This closed reflux uses electricity to regenerate the required chemicals instead of heat as in previous schemes. However, the Solvay reaction may be used to replace the above waste-end reflux. This reaction would be:

The NaHCO_3 would then be treated as described in scheme b) above to close the reflux.

B. Other Separation Processes

The other separation processes investigated in this report are the gaseous diffusion, ion exchange, and gas centrifuge processes. Although the ion exchange and gas centrifuge processes were not regarded as competitive, a brief review of these processes was made for the sake of comparison.

1. Gaseous Diffusion Process

In addition to the chemical exchange methods previously discussed, the production of nitrogen-15 is possible by the gaseous diffusion of nitrogen compounds. In practice, either nitrogen or ammonia would be used. Ammonia has an advantage over nitrogen as a process gas because of its higher separation factor, (ideal α NH_3 = 1.029, ideal α N_2 = 1.018). It should be pointed out that the maximum possible concentration of nitrogen-15 in the product would be 89 per cent using NH_3 and 50 per cent using N_2 without intermediate chemical reactions. The required intermediate chemical reactions would be relatively simple for N_2 , but difficult for NH_3 . In addition, the use of nitrogen would result in lower feed costs and in some savings in construction materials.

Economic optimizations of gaseous diffusion equipment for nitrogen-15 production would probably result in equipment that is considerably different from that used for uranium production. Without going to great detail, several stage sizes have been selected for the ammonia plant. A brief summary of the stage characteristics and costs are shown in Table XI. A schematic layout of the plant is given in Figure 8. Adequate compressors for these stages would have to be designed and developed.

Table XII presents the cost summary for the gaseous diffusion process. The unit cost of product is \$4.60/g. compared to a unit cost of product for the chemical exchange processes estimated as low as \$1 to \$2/g.

2. Ion Exchange Process

The published data of Spedding, et al.⁽³⁵⁾ have indicated that nitrogen isotopes can be separated by ion exchange methods. Calculations by this department⁽³⁶⁾ showed that separation factors and fixed bed HTU values were within range of interest. This observation was confirmed by further data from Spedding⁽³⁷⁾ specifying a separation factor ($\alpha-1 = \psi$) of 0.0255 and a fixed bed HTU of 0.03 inches (for dilute solutions). Although the HTU would be greater with countercurrent systems at somewhat higher flow rates and concentrations, a countercurrent cascade would still appear to represent a reasonable approach.

For the above set of variables, an open reflux countercurrent system was considered which requires NaOH for one reflux and a strong acid such as HCl or H_2SO_4 for the other. Assuming a molar flow of 1.2 times the minimum at the feed point, the cost of purchasing NaOH and H_2SO_4 was estimated to be about \$4 per gram of

product. The cost of chemicals alone for this type of ion exchange system would therefore exceed the total unit cost of chemical exchange processes as low as \$1 to \$2 per gram.

One alternative to such an open reflux system is to recycle the NaCl which is formed. This NaCl may be electrolyzed to form NaOH, H_2 , and Cl_2 and the two gases combined to form HCl. The cost of such a closed reflux system depends upon the operating cost, the initial investment, and the amortization time assumed for the equipment needed. An estimate based upon Blaw Knox type mercury cells indicates that the assumed amortization period would have to be greater than 7-1/2 years to lower the chemical costs below the \$4/g. estimated above. A chemical regeneration scheme such as given in 8a or d above could perhaps lower the chemical costs for ion exchange to the level of chemical costs for closed reflux chemical exchange.

Reflux by NaCl and HCl may be envisioned which allows a relatively simple closed system. Unfortunately, such a system then passes NH_4 -resin countercurrent to highly ionized NH_4Cl and the separation factor of this system for N isotopes is thought to be quite low⁽³⁸⁾ although no experimental evidence has been published.

3. Gas Centrifuge Process

The cost of a plant to concentrate nitrogen-15 by the countercurrent centrifugation of gaseous ammonia has been estimated.

The process was ruled non-competitive when it was found that the plant would cost on the order of \$500,000,000,000 and would consume about twice the present power production of the United States.

It should be remembered that the advantages of centrifugation become evident when dealing with heavy molecules of large mass difference since the separation factor is proportional to the mass difference and not the mass ratio of the molecules.

The centrifuge characteristics and the related costs used are those presented by J. W. Beams, et al.⁽⁴⁰⁾.

BIBLIOGRAPHY

A. Gas-Liquid Chemical Exchange

1. Urey and Greiff, Isotopic Exchange Equilibria, J. Am. Chem. Soc., 57, 321-7 (1935).
2. Urey and Aten, Phys. Rev., 50, 575 (1936).
3. Urey, et al., Concentration of N-15 by a Chemical Exchange Reaction, J. Am. Chem. Soc., 59, 1407-8 (1937).
4. Urey, et al., Concentration of N-15 by Chemical Methods, J. Chem. Phys., 5, 856-68 (1937).
5. Urey and Thode, The Further Concentration of N-15, J. Chem. Phys., 7, 34-9 (1939).
6. Roberts, Thode, and Urey, The Concentration of C-13 by Chemical Exchange, J. Chem. Phys., 7, 137-8 (1939).
7. Urey, The Separation of Isotopes by Chemical Means, J. Wash. Acad. Sci., 30, 277-94 (1940).
8. Nisina, et al., The Exchange Reaction Between Gaseous and Combined Nitrogen, J. Chem. Phys., 9, 571-2 (1941).
9. Norris, et al., The Exchange Reaction Between Gaseous and Combined Nitrogen, J. Chem. Phys., 9, 726 (1941).
10. Joris, The Exchange Reaction Between Gaseous and Combined Nitrogen, J. Chem. Phys., 9, 775 (1941).
11. Stewart, Eastman Kodak Company, Rochester, New York.
12. Clusius, et al., Enrichment of N-15 by Exchange, Chem. Zentr. 1942, II, 1089.
13. Clusius and Becker, Enrichment of N-15 by the Chemical Exchange Procedure, Z. physik. Chem., 193, 64-76 (1943).
14. Thode, Isotope Exchange Factors, Can. J. Res., 23B, 40-7 (1945).
15. Becker and Baumgartel, Enrichment of N-15 in a Two Stage Exchange Plant, Z. Naturforsch. 1, 514-7 (1946).
16. Urey, Report of the Committee on Isotope Separation, WASH-100 (1951).
17. Kirshenbaum, et al., Separation of the Nitrogen Isotopes by the Exchange Reaction Between Ammonia and Solutions of Ammonium Nitrate, J. Chem. Phys., 15, 440-6 (1947).

18. Stewart, Separation of Stable Isotopes, *Nucleonics*, 18 (October 1947).
19. Becker and Vogell, The Shift in Isotopic Abundance in an Equilibrium Solution of Hydrocyanic Acid in Glacial Acetic Acid, *Z. Physik.*, 130, 129-138 (1951).
20. Sugimoto, et al., Concentration of N-15 by Chemical Exchange Reaction Method, *Bull. Chem. Soc. Japan*, 24, 153-5 (1951).

B. Gaseous Chemical Exchange

21. Leifer, The Exchange of Oxygen Between NO and NO₂, *J. Chem. Phys.*, 8, 301-3 (1940).
22. Ogg, Isotopic Nitrogen Exchange Between Nitrogen Pentoxide and Dioxide, *J. Chem. Phys.*, 15, 613 (1947).
23. Taylor and Spindel, Concentration of N-15 in a Gaseous Exchange Column, *J. Chem. Phys.*, 16, 635 (1948).
24. Ogg, et al., Experimental Evidence for the Quasi-Unimolecular Dissociation of Nitrogen Pentoxide, *J. Chem. Phys.*, 18, 573 (1950).
25. Taylor, et al., Annual Progress Report: Separation of Isotopes by Chemical Exchange in Thermal Diffusion Columns, NYO-623, NYO-624, and NYO-625.

C. Gaseous Diffusion, Mass Diffusion, and Sweep Diffusion

26. Wooldridge and Jenkins, Enrichment of Nitrogen in the Isotope N-15, *Phys. Rev.*, 49, 704 (1936).
27. Wooldridge and Smythe, The Separation of Gaseous Isotopes by Diffusion, *Phys. Rev.*, 50, 233-7 (1936).
28. Kruger, Enrichment of N-15 Isotope, *Naturwissenschaften*, 26, 445 (1938).
29. Kruger, Enrichment of N-15 Isotope, *Z. Physik.*, 111, 467-74 (1939).
30. Finneran, J. A., Nitrogen-15 Enrichment in Gaseous Diffusional Equipment, A-3658 (1946).

D. Thermal Diffusion

31. Fleischman, Concentration of N-15 by the Separating-Tube Process of Clusius and Dickel, *Z. Physik.*, 41, 14-8 (1940).
32. Mann, The Thermal Diffusion Constant of Nitrogen, *Phys. Rev.*, 73, 412-3 (1948).

33. Clusius and Becker, The Separation Tube. VI. Isolation of the Mixed Molecule N-14, N-15, Z. Naturforsch, 2a, 154-9 (1947).
34. Clusius, The Separation Tube. IX. Preparation of Pure Heavy Nitrogen-15, Helv. Chim. Acta., 33, 2134-52 (1950).

E. Ion Exchange

35. Spedding, F. H., The Separation of Isotopes by Ion Exchange, Ames Laboratory, April 7, 1954 (ISC-475).
36. Shacter, J., Notes on Ion Exchange, Part II: Preliminary Comments on Ion Exchange for the Separation of N, U, and Pu Isotopes, April 23, 1954 (KD-916).
37. Spedding, F. H., Letter to G. M. Kavanagh dated May 27, 1954, (ORNL-CF-54-5-245).
38. Roberts, J. T., ORNL, private communication.

F. Electrolysis

39. Hutchison, Efficiency of the Electrolytic Separation of Nitrogen Isotopes, Phys. Rev., 75, 1303 (1949).

G. Gas Centrifuge

40. Beams, J. W., NNES X-1, Chapter 11.

H. General

41. Taylor, T. I., verbal information.
42. Worthington, Datin, and Schutz, Ind. Eng. Chem., 44, 910-2 (1952).
43. Hunt, J. Am. Chem. Soc., 54, 3509-12 (1932).
44. Hill and Loucks, J. Am. Chem. Soc., 59, 2094-8 (1937).
45. Seidell, Supplement to the Third Edition, p. 407.
46. Perry, Chemical Engineers Handbook, 3rd ed., p. 596, 610-2, 615.
47. Jones and Pyle, A.I.Ch.E. Symposium Series, Reprint No. 24, December 1953.
48. Arnold, Plank, and Schoenborn, Chem. Eng. Progress, 48, 633 (1952).
49. Perry, Chemical Engineers Handbook, 3rd ed., p. 1457-1558.
50. Chemical Engineering, November 1950, p. 129, August 1953, p. 260-6.
51. Perry, Chemical Engineers Handbook, 3rd ed., p. 250-3.

52. Hougen and Watson, Chemical Process Principles, p. 132.
53. Perry, Chemical Engineers Handbook, 3rd ed., p. 171-2, 250.
54. International Critical Tables, Vol. III, p. 377.
55. Clewett, G. H., Cost of N-15 by Chemical Exchange Process, February 21, 1950.

TABLE I

CHEMICAL EXCHANGE PROCESS BASES COMPARISON

Product Rate - 50,000 Kg./yr. (0.84 lb. mols/hr.) at 99 mol per cent N-15
 Feed Rate - 450 lb. mols/hr. at 0.38 mol per cent N-15
 Waste Rate - 449.16 lb. mols/hr. at 0.19 mol per cent N-15

System	$\text{NH}_3\text{-NH}_4\text{NO}_3$	$\text{NH}_3\text{-(NH}_4)_2\text{SO}_4$	$\text{NH}_3\text{-NH}_4\text{OH}$	$\text{NO-N}_2\text{O}_3$
<u>Reflux Reactions</u>				
Product End	$2\text{NH}_4\text{NO}_3 + \text{Ca(OH)}_2$ $+ \text{Heat} \rightarrow 2\text{NH}_3 + \text{H}_2\text{O}$ $+ \text{Ca(NO}_3)_2$	$(\text{NH}_4)_2\text{SO}_4 + \text{Ca(OH)}_2$ $+ \text{Heat} \rightarrow 2\text{NH}_3 + 2\text{H}_2\text{O}$ $+ \text{CaSO}_4$	$\text{NH}_4\text{OH} + \text{Heat} \rightarrow$ $\text{NH}_3 + \text{H}_2\text{O}$	$\text{N}_2\text{O}_3 + 2\text{FeSO}_4 +$ $\text{H}_2\text{SO}_4 \rightarrow 2\text{NO} +$ $\text{Fe}_2(\text{SO}_4)_3 + \text{H}_2\text{O}$
Waste End	$2\text{NH}_3 + \text{Ca(NO}_3)_2 +$ $\text{CO}_2 + \text{H}_2\text{O} \rightarrow$ $2\text{NH}_4\text{NO}_3 + \text{CaCO}_3$	$2\text{NH}_3 + \text{H}_2\text{SO}_4 \rightarrow$ $(\text{NH}_4)_2\text{SO}_4$	$\text{NH}_3 + \text{H}_2\text{O} \rightarrow$ NH_4OH	$2\text{NO} + \text{O}_2 \rightarrow 2\text{NO}_2$ $\text{NO} + \text{NO}_2 \rightarrow \text{N}_2\text{O}_3$
<u>Supplementary Operations</u>				
1)	$\text{CaCO}_3 + \text{Heat} \rightarrow$ $\text{CaO} + \text{CO}_2$	None	None	$\text{Fe}_2(\text{SO}_4)_3 + \text{H}_2\text{O}$ $\xrightarrow{\text{electricity}}$
2)	$\text{CaO} + \text{H}_2\text{O} \rightarrow$ $\text{Ca(OH)}_2 + \text{Heat}$			$2\text{FeSO}_4 + \text{H}_2\text{SO}_4$ $+ 1/2 \text{O}_2$
<u>Process Variables</u>				
Separation Factor ($\alpha-1$)	0.017	0.015	0.006	0.05
Configuration	Squared Sections $\longrightarrow$			

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TABLE I (Continued)

System	$\text{NH}_3\text{-NH}_4\text{NO}_3$	$\text{NH}_3\text{-(NH}_4)_2\text{SO}_4$	$\text{NH}_3\text{-NH}_4\text{OH}$	$\text{NO-N}_2\text{O}_3$
Length				
Theoretical Stages	1320	1440	3600	440
Ft.	3966	4320	8640	1100
Feed Point Width				
Lb. mols/hr.	15,400	17,400	43,700	6160
Vapor (ft. ³ /hr.)	8,750,000	9,890,000	25,500,000	368,800
Area (ft. ²)	480	540	1257	64
Phases				
Upflow (V)	$\text{NH}_4\text{NO}_3 + \text{NH}_4\text{OH} + \text{NH}_3$	$(\text{NH}_4)_2\text{SO}_4 + \text{NH}_4\text{OH} + \text{NH}_3$	$\text{NH}_4\text{OH} + \text{NH}_3$	$\text{N}_2\text{O}_3 + \text{NO}$
Downflow (V-P)	NH_3	NH_3	NH_3	NO
Contactors	Sieve plate columns	→		
Plate Spacing (in.)	18	18	12	18
Vapor Rate (ft./sec.)	5	5	5.5	1.6
Plate Efficiency (%)	60	60	55	65
Equipment				
Columns				
Number	53	60	116	11
Diameter (ft.)	2.0-12.5	2.0-13.5	4.5-20.0	1.5-9
Height (ft.)	120	120	120	100
Pressure (atm.)	0.67	0.67	0.67	6
Temperature (°F.)	70	70	70	32
Special Design	Top Reflux Towers Kilns	Top Reflux Towers		Electrolytic Cells

TABLE II

MAJOR PLANT EQUIPMENT* - NH_3 - NH_4 NO_3 SYSTEM

<u>Item</u>	<u>Number</u>	<u>Description</u>
Exchange Towers		Sieve plate towers 120 ft. tall having 67 plates each on 18 in. spacing
	11	2.0 ft. diameter
	6	3.5 ft. diameter
	3	5.67 ft. diameter
	3	9.0 ft. diameter
	30	12.5 ft. diameter
Upflow Pumps		Centrifugal pumps - 120 ft. head
	38	15 HP
	4	10 HP
	4	5 HP
	7	2.5 HP
	12	1.5 HP
Blowers		1.67 compression ratio, 7.5 psia. suction pressure
	37	1000 HP - 36,500 cfm.
	4	650 HP
	4	250 HP
	7	100 HP
	12	50 HP
Blenders	7	10,000 gallon capacity
	2	5,000 gallon capacity
Blender Pumps	32	2.5 HP centrifugals, 20 ft. head
N-14 Reflux Towers	4	Sieve plate towers, 12.5 ft. diameter, 120 ft. tall.
Filter	1	Rotary continuous, 1000 sq. ft. effective area.
N-15 Reflux Towers		Sieve plate towers, 80 ft. tall
	1	36 in. diameter
	1	45 in. diameter
	1	74 in. diameter
	1	112 in. diameter
	1	140 in. diameter
	3	180 in. diameter
Evaporator	2	Combined capacity 65,000 gph.

*Type 347 stainless steel construction unless otherwise specified.

TABLE II (Continued)

<u>Item</u>	<u>Number</u>	<u>Description</u>
Heat Exchanger	1	Steel shell and tube, 11,000 sq. ft.
	37	Gas cooler, 10,000 sq. ft.
	37	Gas cooler, 7,000 sq. ft.
	4	Gas cooler, 6,500 sq. ft.
	4	Gas cooler, 4,000 sq. ft.
	4	Gas cooler, 2,500 sq. ft.
	11	Gas cooler, 1,000 sq. ft.
	7	Gas cooler, 750 sq. ft.
	24	Gas cooler, 500 sq. ft.
Rotary Kilns	43	Each 5,000 sq. ft. steel.
Hydrator and Slurrier	4	20,000 gallon agitated tanks.
Refrigeration	2	Each 5,000 ton capacity.

TABLE III
MAJOR PLANT EQUIPMENT* - NH_3 - $(\text{NH}_4)_2\text{SO}_4$ SYSTEM

<u>Item</u>	<u>Number</u>	<u>Description</u>
Exchange Towers		Sieve plate towers 120 ft. tall having 67 plates each on 18 in. spacing.
	9	2.0 ft. diameter
	7	3.0 ft. diameter
	4	4.5 ft. diameter
	3	6.5 ft. diameter
	3	10.25 ft. diameter
	34	13.5 ft. diameter
Upflow Pumps		Centrifugal pump, 120 ft. head.
	42	20 HP
	4	12.5 HP
	4	5.0 HP
	5	2.5 HP
	8	2.0 HP
	10	1.5 HP
Blowers		1.67 compression ratio, 7.5 psia. suction pressure.
	38	1250 HP
	4	750 HP
	4	500 HP
	7	200 HP
	12	100 HP
Blenders	7	12,500 gallon capacity
	2	8,000 gallon capacity
Blender Pumps	36	3 HP centrifugals, 20 ft. head.
N-14 Reflux Towers	4	Sieve plate towers 13.5 ft. diameter by 120 ft. tall.
N-15 Reflux Towers		Sieve plate towers 80 ft. tall.
	2	36 in. diameter
	1	57 in. diameter
	1	74 in. diameter
	1	125 in. diameter
	7	144 in. diameter
Hydrator and Slurrier	4	22,500 gallon agitated tanks.

*Type 347 stainless steel construction.

TABLE III (Continued)

<u>Item</u>	<u>Number</u>	<u>Description</u>
Heat Exchanger (Gas Coolers)	38	12,500 sq. ft.
	38	10,000 sq. ft.
	4	7,500 sq. ft.
	8	5,000 sq. ft.
	4	3,500 sq. ft.
	7	2,000 sq. ft.
	7	1,500 sq. ft.
	12	1,000 sq. ft.
	12	750 sq. ft.
Refrigeration	3	Each 5,000 ton capacity

TABLE IV

MAJOR PLANT EQUIPMENT* - NH_3 - NH_4OH SYSTEM

<u>Item</u>	<u>Number</u>	<u>Description</u>
Exchange Towers		Sieve plate towers 120 ft. tall having 67 plates each on 12 in. spacing.
	30	4.5 ft. diameter
	6	6.25 ft. diameter
	8	8.75 ft. diameter
	6	12.5 ft. diameter
	66	20.0 ft. diameter
Upflow Pumps		Centrifugal pumps, 120 ft. head.
	74	75 HP
	7	15 HP
	9	10 HP
	7	7.5 HP
	31	5.0 HP
Blowers		1.67 compression ratio, 7.5 psia. suction pressure.
	74	2750 HP
	7	1500 HP
	9	1000 HP
	7	750 HP
	31	500 HP
Blenders	16	20,000 gallon capacity
	2	17,500 gallon capacity
Blender Pumps	68	10 HP centrifugals, 20 ft. head
N-15 Reflux Towers		Sieve plate towers, 80 ft. tall
	2	102 in. diameter
	1	144 in. diameter
	17	190 in. diameter
Heat Exchangers (Gas Coolers)	155	15,000 sq. ft.
	157	10,000 sq. ft.
	7	9,000 sq. ft.
	16	7,500 sq. ft.
	31	5,000 sq. ft.
	7	4,500 sq. ft.
	31	3,500 sq. ft.
Refrigeration	4	Each 10,000 tons capacity

*Mild steel construction.

TABLE V

MAJOR PLANT EQUIPMENT* - NO-N₂O₃ SYSTEM

<u>Item</u>	<u>Number</u>	<u>Description</u>
Exchange Columns	6 - 1.5 ft. (dia.) 2 - 3.0 ft. (dia.) 1 - 5.0 ft (dia.) 2 - 9.0 ft. (dia.)	Sieve plate towers each 100 ft. tall having 62 plates on 18 in. spacing.
Pumps	7 3 2 3	0.5 HP, 10.8 gpm. centrifugals 2.0 HP, 43.3 gpm. centrifugals 5.0 HP, 120 gpm. centrifugals 16 HP, 390 gpm. centrifugals
Compressors	7 3 2 3	0.5 HP 1.4 HP 4.0 HP 13.0 HP
NO Burner	1	High pressure reactor approximately 2000 gallon capacity.
Refrigeration System	1	22,040 ton capacity, 0°C.
Agitated Tank	4	4,000 gallon capacity.
Electrolytic Cells	308,000 KW	Individual size and design still require development work. Based on 88×10^6 amps. at 3.5 volts.

*All type 347 stainless steel.

TABLE VI
OPERATIONAL REQUIREMENTS

System	$\frac{\text{NH}_3-\text{NH}_4\text{NO}_3}{3-4-3}$	$\frac{\text{NH}_3-(\text{NH}_4)_2\text{SO}_4}{3-4-2-4}$	$\frac{\text{NH}_3-\text{NH}_4\text{OH}}{3-4-}$	$\frac{\text{NO}-\text{N}_2\text{O}}{2-3}$
Steam (lbs./hr.) $\times 10^{-6}$	0.409	0.187	0.673	---
Water (gpm.) $\times 10^{-3}$	703	450	2910	---
Electric Power (MW)	51.5	67.7	302	308
Refrigeration (tons)	10,000	15,000	40,000	23,000
Gas (Cu. ft./hr.) $\times 10^{-6}$	1.90	---	---	---
Operating Force (men)	670	480	930	460
Feed (NH_3) (Tons/Yr.)	33,500	33,500	33,500	33,500
Inventory Makeup (NH_3) (Tons/Yr.)	11,500	13,000	32,500	4,590
Chemicals				
Ca(OH) ₂ (Tons/Yr.)	--	1,036,000	---	---
H ₂ SO ₄ (100%) (Tons/Yr.)	---	1,372,000	---	---

TABLE VII

COST SUMMARY OF CHEMICAL EXCHANGE PROCESSES FOR N-15 PRODUCTION
AT 50,000 Kg./YEAR

<u>System</u>	<u>NH₃-NH₄NO₃</u>	<u>NH₃-(NH₄)₂SO₄</u>	<u>NH₃-NH₄OH</u>	<u>NO-N₂O₃</u>
<u>5-Year Totals</u> (Millions of Dollars)				
Initial	102.9	82.0	163.7	129.6
Operation	154.8	396.6	293.6	195.4
Total	257.7	478.6	457.3	325.0
<u>Unit Costs of Product</u> (Dollars per Gram)				
Excluding a penalty for deferred production*	1.03	1.91	1.83	1.30
Including a penalty for deferred production**	1.07	2.01	2.34	1.31
Equilibrium Time (Days)	58	86	400	7

*Unit costs excluding a penalty for deferred production were calculated by dividing the total 5-year cost by the amount produced in 5 years at steady state rate of production.

**Unit costs including a penalty for deferred production were calculated by dividing the total 5-year cost by the production realized from the first 5.0 years, i.e., no production during the time required to attain steady state conditions.

TABLE VIII
SUMMARY OF CONSTRUCTION COSTS
(Millions of Dollars)

System	$\frac{\text{NH}_3 - \text{NH}_4\text{NO}_3}{3}$	$\frac{\text{NH}_4 - (\text{NH}_4)_2\text{SO}_4}{4}$	$\frac{\text{NH}_3 - \text{NH}_4\text{OH}}{3}$	$\frac{\text{NO} - \text{N}_2\text{O}_3}{2}$
<u>Installed Equipment</u>				
Columns	11.50	15.31	23.91	0.77
Blowers	2.60	3.21	11.30	0.04
Pumps	0.30	0.21	0.59	0.04
Blenders	0.17	0.19	0.26	----
Filter	0.04	----	----	----
N-14 Reflux Towers	1.63	1.95	----	0.02
N 15 Reflux Towers	1.25	1.56	2.18	0.03
Evaporator	0.38	----	----	----
Heat Exchangers	6.44	8.31	20.70	----
Lime Kilns	14.19	----	----	----
Agitated Tanks	0.11	0.12	----	----
Refrigeration	0.48	0.72	3.26	4.41
Electrolytic Cells	----	----	----	48.13
<hr/>				
Total Installed Equipment	39.09	31.58	62.20	53.48
Piping	17.59	14.21	27.99	16.04
Instrumentation	3.91	3.16	6.22	5.35
Auxiliaries	11.73	7.90	18.66	16.04
Building	3.90	3.90	6.22	5.08
Engineering, Construction, and Contingency	26.68	21.26	42.45	33.60
<hr/>				
Total	102.90	82.01	163.74	129.59

TABLE IX
SUMMARY OF 5-YEAR OPERATIONAL COSTS
(Millions of Dollars)

<u>System</u>	<u>$\frac{\text{NH}_3 - \text{NH}_4\text{NO}_3}{3-4-3}$</u>	<u>$\frac{\text{NH}_3 - (\text{NH}_4)_2\text{SO}_4}{3-4-2-4}$</u>	<u>$\frac{\text{NH}_3 - \text{NH}_4\text{OH}}{3-4-4}$</u>	<u>$\frac{\text{NO} - \text{N}_2\text{O}}{2-3}$</u>
Utilities				
Steam	10.38	4.76	17.08	----
Water	3.70	2.37	15.32	----
Electric Power	19.86	27.70	100.36	101.90
Gas	21.68	----	----	----
Labor (Operating and Maintenance)	16.80	12.10	23.25	11.60
Overhead	16.80	12.10	23.25	11.60
Materials	6.69	5.33	10.64	3.70
Inventory Makeup	4.77	5.39	13.54	1.93
Feed	14.02	14.02	14.02	14.02
Chemicals				
$\text{Ca}(\text{OH})_2$	----	88.19	----	----
H_2SO_4	----	145.30	----	----
Contingencies	40.14	79.32	76.11	50.67
Total	154.84	396.58	293.57	195.42

TABLE X
COMPARISON OF CHEMICAL EXCHANGE SYSTEMS

	<u>Advantages</u>	<u>Disadvantages</u>	<u>Uncertainties</u>
$\text{NH}_3-(\text{NH}_4\text{NO}_3)$ Classical	1) Experimentally demonstrated. 2) Lowest initial cost.	1) Req. large % of nation's capacity HNO_3 . 2) Expensive chemicals - highest operating cost. 3) Special materials of construction due to corrosive chemicals.	1) Variation of separation factor with pressure, temperature, and NH_4NO_3 concentration.
$\text{NH}_3-\text{NH}_4\text{NO}_3$ Closed Reflux	1) Lowered chemical costs. Reduction of large chemical streams.	1) Materials of construction high due to corrosive chemicals. 2) Requires handling of slurries. 3) Extensive auxiliary processes increases plant complexity.	1) Variation of separation factor with pressure, temperature, and NH_4NO_3 concentration. 2) Completeness of product-end reflux not yet demonstrated. 3) Feasibility of regeneration scheme not yet demonstrated.
$\text{NH}_3-(\text{NH}_4)_2\text{SO}_4$	1) Low initial cost. 2) Less expensive chemicals than classical.	1) Relatively high operating cost. 2) Requires handling of slurries. 3) Special materials of construction.	1) Variation of separation factor with pressure, temperature, and $(\text{NH}_4)_2\text{SO}_4$ concentration. 2) Completeness of product-end reflux not yet demonstrated.

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TABLE X (Continued)

	<u>Advantages</u>	<u>Disadvantages</u>	<u>Uncertainties</u>
$\text{NH}_3\text{-NH}_4\text{OH}$	1) Simple reflux systems.	1) Low separation factor. 2) High initial cost. 3) Long equilibrium time. 4) High operating costs.	1) Variation of separation factor with temperature and pressure. 2) Completeness of product-end reflux not yet demonstrated.
$\text{NO-N}_2\text{O}_3$	1) Highest assumed separation factor.	1) Greatest amount of development work required. 2) Operation below room temperature. 3) Leak tightness. 4) Electrolytic type reflux.	1) Value of separation factor. 2) Variation of separation factor with temperature and pressure. 3) Mechanism and completeness of product-end reflux not established (electrolytic cell assumed without specific design or data).

TABLE XI
STAGE PROPERTIES FOR GASEOUS DIFFUSION

<u>Stage Size Designation</u>	<u>Number of Stages</u>	<u>Motor Size, HP</u>	<u>Separation Factor, $\psi = \alpha - 1$</u>	<u>Cost/Stage* M \$/ea.</u>
1	130	20,000	0.022	1,020
2	120	9,700		670
3	60	2,600		220
4	30	1,500		160
5	70	1,125		140
6	120	500		105
7	430	250		70

*Includes total five-year operating costs, as well as equipment and construction costs.

TABLE XII
COST SUMMARY FOR GASEOUS DIFFUSION
(Millions of Dollars)

<u>Item</u>			
Stages and Auxiliaries*			
No. 1	130 required at 1.020 each	132.6	
2	120 0.670	80.4	
3	60 0.220	18.0	
4	30 0.160	4.8	
5	70 0.140	9.8	
6	120 0.105	12.6	
7	430 0.070	30.1	
	<hr/>	<hr/>	
	960	288.3	288.3
Indirect Construction Costs			38.6
Contingencies			49.0
Power, $3,500 \times 10^6$ watts at \$0.005/kw.hr.			763.0
Feed			14
			<hr/>
Total			1152.9**

*Includes total five-year operating costs, as well as equipment and direct construction.

**Unit cost is \$4.60 per gram of nitrogen at 88% N-15 H_3 , 11% N-14 H_2D .

N-15 PRODUCTION BY CLASSICAL CHEMICAL EXCHANGE
OF NH_4NO_3 - NH_3 - SUGGESTED BY UREY AND
PRACTICED BY EASTMAN-KODAK.

Figure 1

N-15 PRODUCTION BY CLOSED REFLUX CHEMICAL EXCHANGE
OF NH_4NO_3 - NH_3

Figure 2

N-15 PRODUCTION BY OPEN REFLUX CHEMICAL EXCHANGE
OF $(\text{NH}_4)_2\text{SO}_4$ - NH_3

Figure 3

N-15 PRODUCTION BY CHEMICAL EXCHANGE
OF $\text{NH}_4\text{OH-NH}_3$

Figure 4

N-15 PRODUCTION BY CHEMICAL
EXCHANGE OF NO AND N_2O_3

Figure 5

*See Table for description of diffusion equipment

N-15 PRODUCTION BY GASEOUS DIFFUSION
OF NH₃

Figure 8