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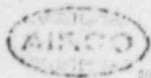
# Noble Gas Recovery Study Maritime Nuclear Ship Savannah

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## FINAL REPORT

NOBLE GAS RECOVERY STUDY  
MARITIME NUCLEAR SHIP SAVANNAH

MARIS 026875

PERFORMED FOR THE ATOMIC ENERGY  
COMMISSION UNDER JOB NO. 2736,  
TODD SHIPYARDS, GALVESTON, TEXAS

By

AIRCO INDUSTRIAL GASES DIVISION,  
AIR REDUCTION COMPANY, INC.,  
NEW YORK 17, NEW YORK

AEROSPACE SYSTEMS  
April 5, 1965

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## SECTION I - INTRODUCTION

This report presents the results of a study performed by Air Reduction for the Atomic Energy Commission under Todd Shipyards Job No. 2736 to determine the feasibility of recovering radioactive purge gas from the nuclear reactor containment compartment on the Nuclear Ship Savannah.

The nuclear reactor on the N. S. Savannah is completely confined within a hermetically sealed containment vessel. This vessel is located in a containment room which is held at a slight negative pressure to insure against any leakage of radioactive material to other areas of the ship. The containment vessel is capable of withstanding the pressure resulting from the rupture of any component inside; however, there is a known leakage rate of about 1-1/2% per day to the containment room.

In the event of a maximum credible accident (MCA), there is a possibility that radioactive Krypton and Xenon could be released. If it were possible to remove or prevent these radioactive gases from being vented, operational benefits could be obtained by reducing or eliminating the "zone of exclusion." The additional fail safe provisions which would be incorporated into the venting system would assure added safety for both ships complement and passengers.

Should there be a control rod failure or other accident which would result in a reactor leak, the radioactive fission gases formed in the nuclear reaction would be confined for a large part within the containment vessel. These gases, however, would gradually leak into the containment room due to the 1-1/2% leakage and eventually be exhausted to the surroundings diluted by about a 200 SCFM air stream. It is estimated that the concentration of fission gases in the containment room can range from 5 to 12 ppm by volume depending upon the activity of the reactor at time of failure.

As a result, Air Reduction was requested to study the problem and:

- 1) develop a system which would reliably remove the fission gases from the exhaust of the ship for up to a two-day period, and
- 2) devise a containment system which would store the fission products indefinitely.

Consequently, several removal techniques were studied so as to arrive at a system which could best meet the needs of the N. S. Savannah. The processes studied incorporated such techniques as adsorption of noble gases by a liquid, adsorption on a solid, or separation from air by cryogenic techniques.



In evaluating these systems, the prime consideration was overall reliability; there must be no compromise on performance. The unit must safely reduce the noble gas concentration in the ship's exhaust to less than 1 part per ten million parts of air. The second consideration is ease of operation since pushbutton starting and unattended operation is envisioned. The third most important consideration was to develop a system which could meet these design requirements at an economical cost.

In addition to determining the practicality of a noble gas recovery system, it is the intent of this report to submit the preliminary engineering details and design criteria necessary for the subsequent preparation of final design drawings and the construction of the proposed unit.



## SECTION II - SUMMARY

### -01 General:

The study was conducted along the outline phases originally proposed by Air Reduction. Slight deviations in the overlapping of tasks was sometimes necessary to permit concurrent development of the program. Where possible, each task was utilized as a building block to form the base for the study and to permit the formulation of a rational system approach.

### -02 Basis for Design:

Prior to the initiation of the Design Criteria and Preliminary Engineering Phases of the study, the system requirements were established. At several joint meetings with representatives of the Atomic Energy Commission, Savannah Technical Staff, Todd Shipyards, and Air Reduction, criteria covering the safety requirements, site location and process flow, etc., were discussed. The quantity, quality, and nature of the radioactive xenon, krypton and compounds of iodine that could be released during an MCA were determined and became part of the system criteria.

These discussions and subsequent correspondence formed the design criteria for the study and is compiled in SECTION III - BASIS FOR DESIGN, of this report.

### 03 Literature Survey:

In order to obtain an information source for system selection, a literature survey was undertaken. Considerable study on the problems of containment and removal of radioactive gases had already been made and therefore only a limited literature survey was required. Though most of the information obtained from the search was not directly applicable to the major problem areas, some valuable guide parameter data was obtained. This information, as well as data and gas purification experience, already available in Air Reduction, gave us the backup to formulate the recovery systems presentation. The literature survey is presented in two major sections of this report; SECTION IV - ANALYSIS OF THE LITERATURE AND APPENDIX B - LITERATURE SEARCH.

### 04 Systems Discussion:

Concurrent with the literature survey, work was started on development of systems to be used for the recovery of the radioactive gases.



Five basic systems, each applicable to the problem, were studied and are described in detail in SECTION V - SYSTEMS DISCUSSION. An additional heat exchanger feature; adaptable to all was also investigated. These systems were then reviewed with regard to performance, safety, operation, and reliability and two were eliminated because of serious deficiencies in one or more of these areas. Detailed cost analyses were not performed since performance, safety, operation and reliability were considered to be of greater importance at that time.

The remaining systems were further analyzed with the element of cost added and two systems were finally selected for comprehensive engineering development and cost analysis. It was determined that these systems would best satisfy the established basis for design.

System C, a cryogenic distillation system shown on Drawing No. 26 8818, presented an attractive choice since Air Reduction has obtained substantial performance data for design from existing operational air separation units. However, a system of this type is unsuitable for quick startup; therefore, the solid adsorption System B with cryogenic enrichment, shown on Drawing No. 26 8817, is selected. This system, besides being the least costly of the two, is simple to operate and extremely reliable. The use of a cryogenic enrichment step reduces the equipment size by increasing the efficiency of the adsorption.

#### 35 Design Criteria:

In SECTION VI - DESIGN CRITERIA, sufficient information is presented to permit the detailed design of the cryogenic adsorption system in a timely and orderly manner.

Process flow conditions are included as well as the type and performance characteristics of all major pieces of equipment; compressors, heat exchangers, storage tank, adsorbent beds, controls, and instrumentation for process monitoring and gas analysis.



## SECTION III - BASIS FOR DESIGN

### 1) General:

This section summarizes the information gathered from Todd Shipyards, the Atomic Energy Commission, and the Savannah Technical Staff that formed the basis for the preliminary design and development of a feasible system for the separation and containment of radioactive noble gases from the N. S. Savannah exhaust air stream.

For clarity, the information is segregated into distinct categories of performance, operation, arrangement, safety and reliability, and shipboard use.

### 2) Performance:

The system shall be designed to reduce the concentration of radioactive noble gases in a 200 cfm air exhaust stream by a factor of 100 or more. For instance, if the inlet air stream contains 12 ppm of radioactive krypton, the system shall reduce the exhaust stream concentration to 0.12 ppm or less.

The system shall be designed to remove, insofar as practical, iodine compounds and iodides from the 200 cfm air stream.

### 3) Operation:

The system shall be designed so that it will be on full time standby, ready for instantaneous operation. Pushbutton actuation is required at the time of an accident. Prior to an accident, the 200 cfm exhaust gas stream shall be bypassed around the system.

The separation and containment of radioactive gases shall be carried out automatically without the need for personnel monitoring and adjustment.

A minimum amount of maintenance shall be required during standby operation.

The liquid nitrogen required for the operation of the system shall be supplied from a shipboard storage vessel. The liquid nitrogen lost due to the vessel heat leak shall be replenished by an existing Norelco liquefier, except that the storage tank shall have sufficient reserve to fully operate the system following a 20-day standby period during which no make-up is supplied from the liquefier.





- (e) The system shall be designed to remove radioactive gases on a continuous basis for two days following an accident.
- (f) The system shall be designed to contain the separated gases for an indefinite period prior to disposal.

3-04 Arrangement:

- (a) The system shall be connected in series with and downstream from the reactor compartment exhaust fan and filter system.
- (b) The shielding required for the system shall be determined and provided by the Savannah Technical Staff. The detail design effort shall identify the areas which have high radioactivity levels and the size of the equipment that requires shielding.
- (c) The design shall provide for an independent pressurized gas supply for instrument and control valve operation.

3-05 Safety and Reliability:

- (a) The design shall provide for the use of redundant equipment where necessary to guarantee adequate reliability.
- (b) The design shall provide for the elimination of the potential leakage of radioactive elements.

3-06 Shipboard Use:

- (a) The equipment shall be designed to accommodate standard shipboard conditions and to fit through a hatch 7'-6" x 7'-6".
- (b) The system shall be designed so that shipboard installation is feasible. The maximum allowable height shall be 8 feet.
- (c) The system shall be operable with a permanent trim of 5°, during a continuous roll of  $\pm 30^\circ$  from vertical (period variable from 14 to 30 seconds), and with a continuous pitch of  $\pm 3^\circ$  from horizontal (7 second period). The approximate accelerations are 0.6g for roll and 0.3g for pitch.
- (d) The noble gas containment bottles shall be unaffected by ship attitude changes including 180° rotation during capsizes.



## SECTION IV - ANALYSIS OF THE LITERATURE

### 4-01 General:

A comprehensive literature survey was conducted to obtain information on the following pertinent subjects:

- . Removal of radioactive noble gases (Krypton and Xenon) from air streams by various methods.
- . Analytical techniques for detecting and quantitatively analyzing the above fission products.
- . Basic materials of construction for containment vessels for radioactive materials.
- . Shipboard installation of liquid air distillation columns.

As was to be expected, the amount of material published in recent years is considerable. Fortunately, since items 1-4 are closely related to the problem of cleaning effluent gas streams from nuclear power installations or of atmospheric decontamination at such locations, some valuable information is found in concentrated form in the proceedings of periodic conferences devoted to air cleaning of AEC installations, nuclear reactor chemistry, analytical chemistry in nuclear reactor technology, etc.

As was also expected, most of the information obtained in the literature search was not directly applicable to the major problem areas. The final system solutions presented are, therefore, based mainly upon the experiences of Airco with similar systems; with the information obtained through the search acting as a reference. The following are abstracts of pertinent literature.

### 4-02 Krypton and Xenon Gas Collection and Containment Systems:

- (a) Method For The Collection and Identification of Radioactive Xenon and Krypton  
J. Flygare, G. Wehmann, A. Harbertson, C. Still  
Presented at Sixth AEC Air Cleaning Conference, Boston, July 7-9, 1959. TID 7593, NSA 15-6245

The Health and Safety Division is responsible for radioactive monitoring of out-plant areas of the National Reactor Testing Station. In order to determine if a leak existed in the Rala off-gas system a sampler capable of collecting radioactive Xenon and Krypton was required so that identification could be made. A sampler consisting of a "U" shaped copper tube filled with activated carbon and cooled with LQN has proved very effective.





Two sizes have been made using 3/4" and 1-1/2" tubing. The collection efficiency for Xenon and Krypton was essentially 100% at flow rates of approximately 1 CFM. Several such samplers were used at strategic locations and disclosed some leaks in the process system. Retention of Xenon and Krypton in activated carbon at room temperature is also discussed.

- (b) The Disposal of Radioactive Fission Gases by Adsorption  
R. Ackley, R. Adams, W. Browning, Jr.  
Presented at Sixth AEC Air Cleaning Conference,  
Boston, July 7-9, 1959. TID 7593, NSA 15-6245

In the operation of various nuclear devices, special provisions must be made for disposal of gaseous fission products to prevent atmospheric containment. A disposal system is described in which noble gas fission products Kr and Xe are delayed by physical adsorption as they pass through an adsorbent such as activated charcoal. A theoretical expression describing this process was developed using a theoretical plate concept and verified experimentally. Retention times were measured for various adsorbents including various grades of charcoal, silica gel, activated alumina, and molecular sieve material. Retention time varies inversely with temperature, and is reduced if CO<sub>2</sub> and water are present. Provision must be made for the dissipation of heat from radioactive decay.

- (c) Xenon - 133 Diffusion On Alumina  
K. Barnes, T. Elleman  
Presented at Second Conference of Nuclear Reactor  
Chemistry, Gatlinburg, Tennessee, October 10, 1961  
NSA 16-26472

Diffusion coefficients were determined for Xe 133 in crystal  $\alpha$  alumina at temperatures ranging from 700 to 900 C. Small spheres of  $\alpha$  alumina were dispersed in a powder and irradiated at low temperatures. These spheres were then separated from the powder and heated at a constant rate. Xe 133 released from the alumina was measured as a function of time. In general Xe133 diffusion is about two orders of magnitude more rapid than oxygen diffusion in  $\alpha$  alumina.

- (d) The Removal of Radioactive Krypton and Xenon  
From a Flowing Helium Stream by Fixed-Bed Adsorption  
W. Graham, D. Morse, Burnette  
Presented at Second Conference of Nuclear Reactor  
Chemistry, Gatlinburg, Tennessee, October 10, 1961  
NSA 16-26472

Fixed-bed adsorption was used to remove Krypton and Xenon from helium streams. The studies were initiated in order to provide the necessary information for the design of a fission-product trapping system for the HTGR. General relationships were derived describing the behavior of radioactive gases in an adsorber. The choice of adsorbents to be tested was limited to various grades of activated charcoal since previous work indicated that charcoal was superior to other adsorbents in this application. The adsorption coefficients for both Krypton and Xenon were plotted against inverse temperature.

- (e) Evaluation of Proposed Krypton-Xenon Adsorption System Designs for the PL-2 Reactor  
 G. Krusen II, and L. Silverman (Harvard Univ., Boston Air Cleaning Lab).  
 August 3, 1962 Contract AT(30-1)-841. 88p.

The proposed design by the Mine Safety Appliance (MSA) Research Corporation of a rare gas handling system for the PL-reactor was examined in detail. This system provides for partial removal of radioactive Xe and Kr from air by low temperature ( $-90^{\circ}\text{F}$ ) adsorption on activated C. The estimated off-gas flow treated is five SCFM. Adsorption is followed by air dilution of the remaining activity. The proposed design was examined to determine whether the AEC-Army Reactor Branch specifications for the resultant activity could be met, and whether the size and weight of the unit could be materially reduced. A parallel search of the literature was made to determine whether other methods could be used to effect the separation. Some were found, but the only one of immediate interest involves condensing the Kr and Xe on activated C at liquid  $\text{N}_2$  temperature. Such a cryogenic system for removing these inert gases from a 2 scfm recirculating flow of  $\text{H}_2$  was installed for operation on the N. S. Savannah. On the basis of available data and calculations, the effluent from the MSA adsorber will not meet the design activity because there is not enough dilution air to attain a level of  $8 \times 10^{-13}$   $\mu\text{c}/\text{ml}$  for  $\text{Kr}^{85}$ . The resultant activity was 38% greater. Added dilution capacity would correct this deficiency. MSA research personnel, in their design considerations, apparently neglected  $\text{Kr}^{85}$  released directly from the fuel elements. Their only concern was with the  $\text{Kr}^{85}$  which is a daughter product of  $\text{Kr}^{85\text{m}}$ . Neither will the effluent meet release criteria if 1% of the equilibrium activity of the PL-2 reactor (7.48 MW) is released to the adsorber over a period of 1 min. while the flow of carrier gas remains undiminished. Recommendations for further research are included. (auth)



- (f) Containment of Radioactive Fission Gases  
by Dynamic Adsorption  
R. E. Adams, W. Browning, R. Ackley  
Oak Ridge Nat. Lab., Tenn. Ind. Eng. Chemistry S1,  
1467-1470 (1954) Dec.

The release of radioactive fission products gases from a circulating fuel nuclear reactor is of such magnitude that direct disposal of these gases was developed. The fission gases Krypton and Xenon, are absorbed from a carrier gas stream onto the surface of a porous adsorbent. Although the adsorption process is reversible, it effectively hinders the passage of fission gases through the system while radioactive decay continually reduces the concentration. Operating parameters which affect the adsorption processes were studied and the data were found applicable for the design of future systems.

- (g) Equilibrium Adsorption of Krypton and Xenon  
on Activated Carbon and Linde Molecular Sieves  
R. Ackley, W. Browning  
NSA 15-17984

The equilibrium adsorption of Krypton and Xenon on Activated Carbon and Linde Molecular Sieves were investigated experimentally. Adsorption isotherms were obtained at 0°C, 25°C and 60°C.

- (h) Adsorption of Krypton and Xenon  
J. Burdick  
NSA 10-6314

The Adsorption isotherms of Krypton and Xenon on Carbon and Silica Gel were measured at temperatures in the range of -183°C to -120°C in static and dynamic systems. The results indicate that either gas can be removed from nitrogen at temperatures below -150°C with a simple fixed bed of activated carbon.

- (i) Adsorption of Xenon in an Activated Charcoal Column  
H. Cantelow  
NSA 13-14267

Performance characteristics of two activated charcoal columns at room temperature in separating fission product Xenon from an air stream were investigated by installing each column in the exhaust from an enclosure in which irradiated slugs were dissolved. Breakthrough curves are presented and the variation in Xenon concentration in the columns is examined. Theoretical treatments of adsorption columns in the literature are found to agree with experimental data. Performance of the columns is



- (n) On the Activities in the Plant Off-Gas  
S. Fuji, J. Manneschildt, T. Mackey, E. Woodall  
NSA 15-17149

Measurements were made of the activities in the laboratory off-gas and efficiencies of the gas cleaning facility were calculated. Sampling was done isokinetically and particulates and adsorbable gases were collected on filter paper and charcoal traps. A special refrigerated system was used for collecting rare gases.  $^{131}\text{I}$  and  $^{106}\text{Ru}$  were the predominant radioactive material in the system. Efficiencies of the gas cleaning system ranged from less than 50% for the gaseous material to more than 99.9% for the particulates.

- (o) Adsorption of Xenon on Activated Charcoal  
W. Kenney, A. Eshaya  
NSA 16-14814

Dynamic adsorption experiments were run to determine the shape and position of breakthrough curves for Xenon from Xenon-Helium mixtures. Flow rates over the range of 500 to 3000 CC per minute were put through an activated charcoal bed 1.38 inches by 10.5 inches. Inlet concentrations varied from 0.02 to 1.0% Xenon.

- (p) Retention Efficiencies of Selected Adsorbents for Krypton  
R. Koch, G. Crandy  
NSA 14-11370

An investigation was conducted to determine the retention efficiencies of selected adsorbents for Kr in a stream of nitrogen. The adsorbents studied were carbons, mineral, and inorganic compounds.

- (q) Adsorption of Krypton and Xenon By Various Materials  
M. Lloyd, R. McWees  
NSA 16-3068

The adsorptive capabilities of various inorganic adsorbents and activated charcoals for Krypton and Xenon were determined. Columbia-G activated carbon had the highest capacity for both Krypton and Xenon at pressures between 0.01 and 125 mm Hg, and temperatures between 2 to 85°C. The capacity of molecular sieves (5A or 11X) ranged from 11 to 20% of the Columbia G. Both the carbon and molecular sieve adsorbed 11.5 times more Xenon than Krypton. Also, the presence of water reduced the capacity for Krypton.

- (r) Adsorption of Radioactive Gases on Activated Carbon  
 R. Madey, J. Barker, M. Beebe, Stephenson  
 NSA 15-1941

An experiment was conducted to study the characteristics of a carbon adsorbent bed, receiving radioactive inert gas in a helium stream.  $Kr^{85}$  was used since it can be readily produced and detected. The experiment was designed so that the adsorber saturation time was comparable to the mean  $Kr^{85}$  lifetime. The concentration was on the order of  $10^{-12}$  atom fraction.

- (s) An Absorption Process for Recovery of Fission Product  
 Noble Gases  
 M. Steinberg, B. Manowitz

Because of the possible pollution of the atmosphere by the accumulation of radioactive gaseous fission product Xenon and Krypton from expanding nuclear operations and because these substances are potentially valuable, a need arises for their removal, containment, and recovery. Recovery by adsorption on charcoal is reviewed. Mainly because of the safety hazards involved, a liquid absorption system presents several advantages. Experimental data on the solubility of Xenon and Krypton in a number of solvents have been obtained by a modified McDaniel procedure and by radioactive tracer techniques. Certain liquid solvents have been found to have a significant affinity for Xenon and Krypton. Solubility data are correlated in terms of the solvent cohesive energy density and found to conform to Hildebrand's solubility theory. The solubility of Xenon and Krypton in a suitable liquid solvent, at temperatures ranging from room temperature up to  $150^{\circ}C$ , and with gas compositions ranging from pure gas to  $\approx 10$  ppm by volume in nitrogen shows a decrease with increasing temperature and demonstrates the applicability of Henry's law. Rates of absorption have been determined by the operation of a continuous counter-current packed absorption column. Under the conditions of the experiment, the rate of absorption of the noble gases appears to be liquid film controlled and is expressed as a function of the solvent mass flow rate. Based on the experimental data, the design of an absorption-stripping process for the removal and concentration of xenon and krypton from a fuel processing plant is presented.

- (t) The Recovery of Fission Product Xenon and Krypton by  
 Absorption Processes  
 M. Steinberg  
 NSA 13-13329

Highly concentrated fission product xenon and krypton can be economically produced with high rates of recovery from diluted nuclear off-gas streams by means of a continuous selective solvent





absorption process. The process involves a three-column absorption-fractionation-stripping operation. The absorber removes xenon and krypton along with part of the diluent gases, nitrogen and oxygen; the fractionator removes the nitrogen and oxygen from the solvent; and the stripper produces the highly concentrated noble gas product. Solvent flow rates and stripping gas requirements are considerably reduced by operating the absorber under higher pressure, and the fractionator and stripper at lower pressures.

Experimental data on the solubilities of xenon, krypton, nitrogen, oxygen, and argon in liquid nitrous oxide and Freon, measured over a range of temperatures, are presented. The selectivity and solubility of xenon and krypton in these solvents at temperatures of the order of  $-70^{\circ}\text{C}$  are significantly higher than in solvents at ambient temperatures. Based on these and earlier solubility measurements, process designs, flow sheets, and column design data are given for each of three solvent systems: kerosene, liquid nitrous oxide, and liquid Freon.

The kerosene-carbon dioxide stripping process has the advantages of operation at ambient temperatures and elimination of all heat exchange equipment; however, the cost of carbon dioxide is an economic factor in this case. In the liquid nitrous oxide process, the necessity for a nitrous oxide removal operation is eliminated, but the hazard of using a thermodynamically unstable solvent must be taken into consideration. The Freon process has the lowest refrigeration requirements, a low solvent cost and maximum stability.

The use of solvents having boiling points higher than those of xenon and krypton has the advantages of reducing equipment freeze-up, shielding requirements, and accumulation of hazardous materials in the process.

- (u) Recovery of Fission Product Krypton From Nuclear Fuels  
R. Taylor  
Ind. Chemist 39(7) 358-64-CA59, 14847

The development of a process for recovering  $\text{Kr}^{85}$  from off-gases resulting from the dissolving of spent nuclear fuel elements as described. Processes for concentrating and purification of the recovered gas are also described.

- (v) Recovery of Fission Product Noble Gases  
M. Steinberg, B. Manowitz  
Ind. Eng. Chemistry 51:47-50-NSA 13-4491

This paper deals with the development of an absorption-stripping process for the concentration of xenon and krypton from a fuel-



processing plant. Tests were conducted using kerosene-base solvents, i.e., AMSCO 123-15 (American Mineral Spirits Solvent Company No. 140).

Based on rates of absorption and other tests conducted, a system of noble gas removal and recovery was developed to process off-gas effluent from a 1-metric ton per day fuel processing plant.

- (w) Systems for Handling Radioactive Gases  
J. Weisman, M. Griffet  
CA 59, 71359

Radioactive isotopes of Xe and Kr are adsorbed by charcoal, and held until the radioactivity has decayed sufficiently to allow the gases to be discharged safely into the air. In this method, several beds of charcoal are used to retain the radioactive Xe and Kr.

- (x) The Adsorption of Krypton and Xenon on  
Activated Charcoal and its Uses in Research  
and Industry  
L. Weller  
NSA 13-18899

A bibliography on the adsorption of Krypton and Xenon on activated charcoal covering the period between 1905 and 1959 is presented. In addition, references on their uses in research and industry are also included.

- (y) Measurement and Analysis of the Holdup of Gas Mixtures  
By Charcoal Adsorption Traps  
W. Browning, C. Bolta  
NSA 10-8505

The holdup behavior of gaseous fission products in charcoal traps was investigated as a function of trap geometry, type and amount of charcoal, trap temperature, flow rate and type of inert gas used as a carrier. An analytical expression is presented for the calculation of holdup curves for gases when trap geometry and adsorption isotherms for the gases or charcoal are known.

- (z) Removal of Fission Product Gases From  
Reactor Off-Gas Streams by Adsorption  
W. Browning, R. Adams, R. Ackley  
Presented at American Nuclear Society Meeting, Detroit,  
Michigan, December 10, 1958.

A disposal process is described in which the noble gas fission products, krypton and xenon, are delayed relative to the sweep gas by physical adsorption as they pass through an adsorbent such as activated charcoal. A theoretical expression describing this process was developed, using a theoretical plate analysis and was





verified experimentally. The retention time for a gas present in trace concentration is proportional to the amount of charcoal in the adsorber bed and to the adsorption coefficient which is evaluated experimentally for a particular combination of materials and conditions. The retention time is inversely proportional to the volume flow rate of the sweep gas. The retention times of experimental adsorbers have been measured by a radioactive tracer technique using krypton-85 and xenon-133 to typify fission gases. Retention times were measured for various adsorbents including different grades of activated charcoal, silica gel, activated alumina, and molecular sieve materials. Activated charcoal was the most effective adsorbent.

Retention times were measured using helium, hydrogen, argon, nitrogen, oxygen, air, CO<sub>2</sub>, Freon-12, and krypton as sweep gases, individually. Water vapor reduces the retention time for krypton on charcoal as does CO<sub>2</sub>. Krypton interferes negligibly up to 1000 microns partial pressure. Provision must be made for the dissipation of heat from radioactive decay. Retention time decreases logarithmically as temperature increases. Ignition temperatures in oxygen were determined and methods for prevention and control of charcoal fires were investigated. Experimental results described were used to design off-gas adsorber systems for in-pile experiments and to analyze the performance of an adsorber for a homogeneous circulating fuel reactor. These adsorber systems performed satisfactorily for times in excess of one year.

4-03 Analytical Techniques for Detecting Small Quantities of Radioactive Gases

- (a) A Continuous Stripper For The Determination of Dissolved Gases and Fission-Product Gases Presented at Fourth Conference of Analytical Chemistry in Nuclear Reactor Technology  
TID 7606-NSA 15-19270  
S. P. Gibson, G. M. Allison, and J. F. Atherley  
(Atomic Energy of Canada Ltd., Chalk River, Ont.)

A gas stripper device and procedure are described for making continuous, semi-continuous, and spot determinations of dissolved gases in high-temperature and pressure water systems. Two of the gases, N<sub>2</sub> and H<sub>2</sub>, are determined by gas chromatography, using a Vapor Fractometer. The concentrations of Xe<sup>133</sup>, Xe<sup>138</sup>, and Kr<sup>88</sup> in the gases are determined by gamma spectrometry. The techniques involved in these analyses are discussed. Data are presented to show that 95% of the N<sub>2</sub> and H<sub>2</sub> plus 90% of the fission-product gases are removed by this continuous stripping process.



- (b) The Significance of the Air Cleaning Problem  
in Aircraft Nuclear Propulsion  
R. Baker  
Presented at Sixth A.E.C. Air Cleaning Conference  
July 7-9, 1959 at Boston NSA 15-6245

The H.T.R.E. and its off-gas monitoring and disposal system are discussed.

- (c) The Determination of Trace Elements In  
Reactor Materials by Methods of Neutron  
Activation Analysis  
G. Leddicotte, W. Mullins, L. Bate, J. Emery  
Presented at 1st Conference of Analytical Chemistry  
in Nuclear Reactor Technology - Gatlinburg, Tennessee,  
September 9, 1958  
NSA 12-16223

Neutron activation analysis is used to determine trace elements in such reactor materials as ores, structural materials, cooling water, and moderators.

- (d) Analytical Requirements on the Lockheed Critical  
Experiment Reactor of the Radiation Effects, and of  
Radioactive Waste Disposal  
J. Edgerton  
Presented at the 2nd Conference of Analytical Chemistry  
in Nuclear Reactor Technology, Sept. 9, 1958 - Gatlinburg, Tenn.  
NSA 13-12445

The analytical chemistry requirements of the Lockheed Critical Experiment Reactor are discussed. The use of process instrumentation and the application of analytical chemistry for maintaining proper conditions for operating the reactors. Methods for the analysis of reactor materials including corrosion, activation, and fission products are discussed. The radioactive waste disposal system for abnormal and normal operation of the reactor is described.

- (e) Continuous Analysis of Radioactive Gas and Liquid Streams  
S. McEwen  
Presented at the Fourth Conference of Nuclear  
Reactor Technology. October 12-14, 1960 - Gatlinburg, Tenn.  
NSA 15-19270

Instrumentation for continuous analysis is discussed. In gas and liquid-stream analysis, sampling and measuring techniques vary widely in most instances. There are some methods of analyzing two types of streams which do overlap in type of instrumentation



but only in rare instances and in technique as well. The instruments, for continuous gas and liquid analysis include on-stream analyzers for measurement of components in the following ranges: percentage, parts per million, and parts per billion. The techniques used in each of these ranges are described. In general, those methods of analysis are emphasized which deal with the determination of components in the ppm and ppb range. The analytical techniques cover measurement of gases in gas streams, gases in liquids, liquids in liquids, and dissolved solids in liquids.

- (f) An In-Cell Gamma Analyzer  
V. Upson, F. Roberts  
Presented at the Fifth Conference of  
Analytical Chemistry In Nuclear  
Reactor Technology  
Oct. 10-12, 1961 - Gatlinburg, Tenn.  
NSA 16-23613

A close-coupled gamma spectrometer for viewing samples inside the hot cell was utilized to provide analytical control in the Hanford Laboratories' High-Level Radiochemical Facility, yielding rapid and accurate isotopic analyses of undiluted samples. Design criteria, based on an expected maximum cell activity of 100 kilocuries of Ce-Pr<sup>144</sup> (2.2 Mev) and a sample activity range from 0.2 to 2000 mc/ml were exceeded. At twice the anticipated maximum cell activity (~1000 r/hr at the sample station) it was possible to detect as little as 0.01 mc/ml and to evaluate 0.1 mc/ml isotopic activity in the sample; sample activities exceeding m curies/ml can be tolerated. Two through-the-wall access tubes were utilized to accommodate two detectors, each of which views two sample positions. The two positions for each detector were designed to yield geometries differing by a factor of 10, and the two detectors were collimated to differ by about 100, yielding four sample positions in approximate decade steps. Pulse-height analysis of the detector outputs is performed by a 400-channel analyzer operating at 1.00 Mev per 100 channels, and utilizing instrumental background subtraction to yield direct printout of the gamma spectra. Isotopic analysis usually requires only a 1 or 2-minute counting period. The instrument proved to be an extremely valuable and reliable tool in both research and chemical processing operations. (auth)

- (g) Determination of the Distribution of Fission Xe  
in Irradiated UO<sub>2</sub> Fuel Elements  
W. Morgan, R. Hart, F. Miller, W. Olmstead  
Presented at the Third Conference of Analytical  
Chemistry in Reactor Technology  
October 26, 1959 - Gatlinburg, Tennessee  
NSA 15-8732



In the development of  $UO_2$  fuel elements for power reactors, one of the important aspects to be considered is the physical behavior of the fission gas. The engineering concern is mainly that of pressure build-up inside the sheath, due to the release of fission gas from the oxide. Two analytical methods in use at Chalk River have contributed considerably to the investigation of the fate of the fission gas in experimental fuel elements; measurement of xenon released on puncturing the sheath of the fuel element and measurement of the radial distribution of xenon remaining in the  $UO_2$ . These two methods are described. (auth)

- (h) A quantitative Determination of Several Short-Lived Iodine, Barium, and Strontium Fission Products in Gas Cooled Reactor Effluents  
L. Reed, M. Myers, W. Sabal  
Presented at the Third Conference of Analytical Chemistry in Nuclear Reactor Technology  
Oct. 26, 1959 Gatlinburg, Tennessee  
NSA 15-8732

Gamma scintillation spectrometry was used as a basis for the quantitative determination of absolute disintegration rate values for  $I^{131}$ ,  $I^{132}$ ,  $I^{133}$ ,  $I^{134}$ ,  $I^{135}$ ,  $Ba^{139}$ ,  $Ba^{140}$ ,  $Sr^{91}$ , and  $Sr^{92}$  collected from reactor effluent air on activated coconut charcoal. After chemical separations for the various elements were made, the complex spectra resulting were treated mathematically, resulting in the solution of simultaneous linear equations to yield d/m values for the specific isotope. Corrections were made for any absorber between the source and NaI (Tl) crystal, for the total absolute detection efficiency for the counting geometry used, for the peak to total ratios for the particular photopeak, for the decay scheme, and for any internal conversion, thus obviating the need for standard plates of the isotope in question. The method is applicable to  $\gamma$ -emitters in general, providing values for corrections to be applied are available.

- (i) Fission-Product Analysis of Fast Reactor Plutonium Fuels  
J. Meadows, G. Matlack, G. Nelson  
Presented at Third Conference of Analytical Chemistry in Nuclear Reactor Technology  
October 26, 1959. Gatlinburg, Tennessee.  
NSA 15-8737

For the purpose of evaluating the pyrometallurgy program at Los Alamos for the reprocessing of fuels from fast reactors, it was necessary to determine fission products in the presence of large amounts of plutonium, especially when low burn-up fuels were to be analyzed. From results of a testing program in which classical analytical methods originally developed for uranium



fuels were used to determine fission products, it was shown that adequate decontamination from plutonium and americium was not achieved in a number of cases. New or modified methods were developed for Zr, Nb, Ru, Rh, Ce, Nd, and Pr. All procedures were designed to utilize sample aliquots containing as much as 50 mg of plutonium and to give  $\alpha$  decontamination factors of not less than  $10^5$  and as high as  $10^9$ , when from 10 to 50 mg of carrier were added. Ion-exchange methods proved to be the most effective for removing plutonium and americium in those cases where these elements caused interference. Methods for 14 fission products were tested or devised for use in the analysis of plutonium fuels.

- (j) Separation and Estimation of Krypton and Xenon by Gas Chromatography  
R. Anbeau, L. Champeix, and Reissy  
Journal of Chromatography

The application of chromatograph principles to the separation and quantitative estimation of the fission gases krypton and xenon.

- (k) A Gross Gaseous Particulate Fission Product Monitoring System  
F. Boone  
Journal of American Industrial Hygiene Association  
24 (6): 611-17. CA 60,10162g.

A moving filter paper device with anthracene scintillation crystals followed by a detection chamber with a single sodium iodide scintillation crystal gives a continuous detection of the fission product release from an open gas cooled reactor.

- (l) A New Method for the Determination of Some Gaseous Fission Products  
A. Bouville, D. Blanc, J. Couly, J. Fenton  
NSA 18-9993

Conventional methods for the determination of naturally radioactive gases were adapted to the detection of artificial radioactive gases having a chain of solid descendants. The isotopes of Kr 88 to 97 were the fission products studied.

- (m) On-stream Radioactivity Monitor for Gas Handling Systems  
Rev. Sci. Instr. 31:786 CA 55,21688h

Designed for use in the development and testing of a gas chromatography system for the separation of Xenon and Krypton, the new system continuously monitored the concentration of radioactivity





in a dynamic gas system. It has been used for measuring Kr<sup>85</sup> and Xenon 133 in several types of gas tracer experiments. The detector unit consisted of a gas flow chamber, a scintillation head, and associated electronic apparatus and was an integral part of the system. It could be used with vacuum or pressurized operations and for the measurements of  $\beta$  or  $\gamma$  radiations.

- (n) A Simplified Method for Radio Iodine Analysis  
H. Eiland  
Third Conference Analytical Chemistry  
In Nuclear Reactor Technology, Gatlinburg, Tennessee  
October 26, 1959  
NSA 15-8732

A radio-chemical method of analysis for gross iodine activity in the primary coolant of pressurized water reactors was developed at KAPL. It is based upon the rapid isotopic exchange which occurs under suitable conditions between fission product iodine and the iodine atoms in preformed silver iodide. The recovery of iodine was shown to be  $88 \pm 5\%$ . The procedure was used extensively at KAPL for application when speed and simplicity were of greater importance than accuracy.

- (o) Krypton Solubility  
R. Keeler, C. Anderson, S. Klach, R. Chappel  
NSA 13-19034

The solubility of krypton under reactor conditions to provide a basis for estimating the concentrations and locations of fission product rare gases in a homogeneous reactor was investigated. In addition selective chemical and physical properties of iodine were studied to predict the behavior of iodine in the reactor.

- (p) Movement of Highly Radioactive Gases in Absorption Tubes  
E. Glueckauf  
Ann. N. Y. Acad. Science 72:562-91  
CA 53,14602i

The gas chromatography of these gases is materially different from that of other adsorbates owing to heat effects from radiation adsorption. The breakthrough volumes and times through externally cooled charcoal columns, the radial temperature distributions and the heat to be removed at various points along the column were studied for mixtures of radioactive xenon and krypton. An estimate of channeling effects owing to high interior temperatures were made. Elution in insulated columns were also studied. When the ratio of radioactive power to gas flow approaches 8 joules per M<sup>3</sup> separation is no longer possible.

- (q) Limits of Sensitivity in Monitoring Radioactive Gases with Particular Reference to Krypton 85  
 J. Shapiro, R. Yoder, L. Silverman  
 Presented at Eighth A.E.C. Air Cleaning Conference,  
 Oak Ridge, Tennessee Oct. 22-25, 1963  
 NSA 14125

Calculations of efficiencies of idealized ionization chambers and particle detectors for monitoring radioactive gases are presented. Kr <sup>85</sup> was used as the radioactive gas in the calculations and measurements. It is shown that for levels of the order of  $10^{-7}$   $\mu\text{C}/\text{cc}$ , a simple cylindrical G-M counter is adequate if a large volume can be viewed, and it is the simplest detector available. (M.C.G.)

4-04 Radioactive Iodine and Iodide Collection Systems

- (a) Iodine Collection Studies  
 L. Silverman, R. Dennis, E. Kristal, F. Stein  
 Presented at Sixth AEC Air Cleaning Conference  
 Boston, July 7-9, 1959. NSA 15-6245

Several methods for the removal of radioactive iodine from process gas streams are under investigations. Test objectives to develop a system which will afford at least 90% I<sup>131</sup> collection. Also, to have low pressure drop, be inexpensive, simple to fabricate and maintain, long life, good corrosion resistance, function at temperatures up to 300°C, and be non-combustible. Data is presented for several systems.

- (b) Removal of Iodine From Gas Streams  
 R. Adams, W. Browning  
 Presented at Seventh AEC Air Cleaning Conference,  
 Brookhaven, New York October 10-12, 1961  
 NSA 16-29214

The efficiency of activated charcoal silver and copper surfaces for adsorption of iodine vapor was studied under various conditions. Iodine vapor is removed from air at 25°C by activated charcoal with efficiencies as high as 99.999+%. Copper and Silver plated copper ribbon exhibit efficiencies up to 98 to 99%.

- (c) A Review of Iodine Collection Studies  
 R. Dennis, L. Silverman, F. Stein  
 Presented at Seventh AEC Air Cleaning Conference  
 Brookhaven, New York. October 10-12, 1961  
 NSA 15-6245

Results of past Iodine Collection Studies are reviewed. Performance data not previously reported are given for metal and mineral collectors under a variety of operating conditions.



Studies conducted by Harvard Air Cleaning Laboratory under AEC contract.

- (d) Air Cleaning Studies at Harvard University  
L. Silverman  
Presented at Fifth AEC Air Cleaning Conference  
June 24-27, Boston, Massachusetts  
NSA 12-12147

Several projects are mentioned which are covered by the papers following this one. The use of slag wool coated with Ag for I<sup>131</sup> removal is discussed. The efficiency of slag wool filters on fly ash was investigated and data are tabulated.

- (e) Design Considerations for Exhaust Systems  
Involving Radioactive Particulates  
A. Fuller  
Presented at Eighth AEC Air Cleaning Conference  
Oak Ridge, Tennessee. October 22-24, 1963  
NSA 18-14116

The dependability required of exhaust ventilation systems handling radioactive particulates makes it essential that through investigations concerning needs be made and good engineering practices be applied. A number of designer "short comings" are outlined with references.

- (f) Radioiodine Adsorption Systems for the N. S. Savannah  
W. Browning, R. Adams, W. Johnson  
Included in Reactor Chemistry Division  
Annual Progress Report for period ending January 31, 1963  
NSA 25087

Studies of the removal of radioiodine from steam-air mixtures continued in support of the NS "Savannah" program. Over 60 small-scale tests and 14 large-scale tests were completed. Activated-charcoal units, prepared in the same manner and using materials similar to those employed in the units installed on the NS "Savannah" were utilized in the large-scale laboratory tests. These tests, conducted with continuous I<sub>2</sub> injection at 96 to 100°C and with 80 to 90% saturated steam in air, showed the efficiency of the charcoal unit to be  $(99.86 \pm 0.07)\%$  at the 95% confidence level. In-place tests of the full-scale filter-adsorber units in the reactor compartment ventilation systems of the NS "Savannah" were conducted with radioactive I<sup>131</sup> and non-radioactive I<sup>127</sup>. The emergency ventilation system, greater than 99.9%, whereas the main ventilation system, containing no charcoal, showed I<sub>2</sub>-retention efficiencies ranging from 90 to 98% in several series of tests.



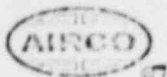
- (g) Characterization and Control of  
Accident Released Fission Products  
W. Browning, R. Ackley, M. Silverman

Included in Reactor Chemistry Division Annual  
Report for period ending January 31, 1963  
NSA 25084

Two methods of determining the form of radioactivity in gases were investigated. Diffusion coefficients of small particles and of radioactive vapors such as  $I_2$  are determined by measuring the distribution of radioactivity on the walls of a channel previously exposed to gas carrying radioactive materials and flowing under laminar conditions. This technique was employed to demonstrate that high-efficiency, low-pressure-drop filters removed about 10 to 75% of the activity from air streams carrying  $I^{131}$  adsorbed on 0.004- $\mu$ -diameters  $Al_2O_3$  particles. Beds containing a 0.75-in. depth of -6 + 16 mesh activated C removed essentially all the  $I_2$  vapor but only 75 to 90% of the 0.003- to 0.006- $\mu$   $Al_2O_3$  particles, the higher efficiency being associated with the finer particles. Fog-condensation and foam-encapsulation methods removed 85 to 99% of these very small particles. The second method of measuring the size of radioactive aerosols makes use of a filter having a uniform fiber diameter to permit theoretical analysis and having a layered structure to facilitate separation of the fiber bed into discrete layers for radioassay after exposure to the aerosol. Preliminary results obtained with a radioactive aerosol of 0.004- to 0.03- $\mu$  particles labeled with  $Zn^{65}$  indicate that the techniques used are satisfactory.

- (h) Behavior of Radioiodine  
Oak Ridge National Laboratory, Tennessee  
Reactor Chemistry Division Annual Progress Report  
for period ending January 21, 1964. ORNL-3591  
NSA 22111

An investigation was made of the origin, identity, and behavior of iodine compounds which appear when air and iodine vapor are mixed. Information is needed on these points in order to design radioiodine removal systems. Rates of deposition of iodine compounds were measured by use of successive diffusion tubes having surfaces of silver, rubber, and activated carbon. Elemental iodine deposited on the silver with its characteristic diffusion coefficient of about 0.08  $cm^2/sec$ ; a compound or group of compounds deposited on the rubber to give an indicated diffusion coefficient of about 0.05  $cm^2/sec$  suggestive of a molecular weight of the order of 300 or higher; other iodine materials deposited on the activated carbon to yield a diffusion coefficient of 0.10  $cm^2/sec$ ., implying a molecular weight somewhat lower than that of  $I_2$ . This behavior was typical of iodine from two greatly differing sources. As the iodine concentration in



air decreased, the proportion of iodine present in forms other than molecular iodine increased. The sorption characteristics of fission products are of significance in predicting their deposition behavior in flowing gas systems. The adsorption of iodine on commonly used stainless steels was explored with metal temperatures ranging from 24 to 700°C and with iodine vapor concentrations varying from 0.0004 to 2200 mg/m<sup>3</sup>. Iodine vapor was found to react chemically with stainless steel surfaces to form metal iodides. The capacity of the surface varied with the condition of the adsorbing surface. Passivity towards iodine adsorption was frequently observed after a small amount of material was sorbed. The kinetics of the chemisorption process also varied, apparently with changing surface conditions, even during the course of a single experiment. The iodine sources used were analyzed for forms other than I<sub>2</sub>.

Methyl iodide was found, but it constituted less than 0.1% of the total iodine. Fractional sublimation of the iodine sources in vacuum ensured that only I<sub>2</sub> was used in the adsorption experiments. Charcoal traps remove molecular iodine from gases with a high degree of efficiency, but they are susceptible to destruction when exposed to oxidizing gases at high temperatures, conditions which could conceivably exist in a reactor accident. Preliminary experiments to explore alternate trapping materials indicated that a bed of platinized alumina (a hydroforming catalyst) would serve only to delay the passage of iodine when the bed is exposed to flowing air at temperatures above 400°C. Trapping of radioiodine by high-temperature exchange with non-radioactive temperatures at which significant oxidation of the inorganic iodine occurs (600°C or higher).

- (i) Proposed Method for Removal of Radioiodine Vapor from Experiment Off-Gas System of the ORR  
R. Adams and W. Browning  
NSA 12-10488

Various methods of removing radioiodine from air streams were surveyed for possible use in removing radioiodine released from reactor experiments into the off-gas system. Solid adsorber materials appear best suited for this purpose. An iodine decontamination factor of 2,000 was experimentally determined for activated charcoal, and Linde Molecular sieve (13x) using I<sup>131</sup> to simulate mixed radioisotopes of iodine. A conceptual design for an iodine trap containing activated charcoal is presented. The iodine removal efficiency from a hypothetical trap is discussed under conditions resulting from the discharge of 3.0 x 10<sup>4</sup> curies mixed radioisotopes of iodine.



- (j) Removal of Radioiodine from Air-Stream Mixtures  
R. Adams, W. Browning  
NSA 15-19449

The removal of radioiodine vapor from air-stream mixtures such as those from a nuclear incident in a pressurized water reactor was investigated. Activated charcoal traps, simulating part of a commercial charcoal cannister were tested at gas velocities of 23.9 to 74.9 ft/min. over the temperature range of 75°C to 118°C. The iodine removal efficiency was found to range between 99.80 to 99.94%. It was reduced to 99.54% in a test run with a gas velocity of 290 ft/min. at 105°C.

- (k) The Removal of Iodine from Gas Streams  
By Reaction with Silver in Packed Towers  
R. McNabney, A. Lyon

A process was developed for the removal of iodine alone in the presence of the various other constituents of pile dissolver off-gases. The principal feature of the process is the use of a column packed with berl saddles coated with silver nitrate or silver. Experiments were conducted with a 2" diameter 17" long packed column through which a stream of air at 2 cfm containing NO<sub>2</sub>, water vapor, and iodine was passed. This tower is equivalent to a 6' plant scale unit operating at Hanford.

- (l) Laboratory Plate-Out Study of Iodine  
N. Myers  
NSA 15-32974

A study was undertaken to provide information on the quantitative plate-out, a deposition of fission products on hardware and effluent sampling lines in the various reactor systems. The effect of temperature, flow, bends, and fittings were studied.

- (m) Application of Activated Carbon  
in Reactor Containment  
G. Prigge  
NSA 17-4232

An activated-carbon bed or filter was designed to remove 99.9+% of the radioactive halogen vapor that could be released as a result of a containable accident of a water cooled and moderated reactor. The unit was tested over simulated conditions of normal exposure of the carbon filters to the air in the ventilation exhaust as well as to exposure conditions postulated for a containable reactor accident. The tests indicated a 99.99+% efficiency when exposed to air, dry steam and mixtures of steam, air, and entrained liquid water particles.



- (n) A Short Laboratory Investigation of  
Radioiodine Plate-Out  
G. Tuck, M. Myers

The removal and plate-out of radioiodine was studied by blowing hot air through a short section of 3/8" stainless steel tubing containing plated-out fission products and then through a 30-foot clean 3/8" tube. The removal of I by the air at various temperatures, and subsequent plate-out in the clean tubes, were measured.

- (o) Removal of Radioiodine from Air Streams  
By Activated Charcoal  
R. Adams, W. Browning  
NSA 14-11371

The efficiency of activated charcoal for adsorption of iodine vapor from air streams were measured by using a radioactive tracer method. Efficiencies of 99.6 to 99.999+% were obtained at various operating conditions. Various materials were considered for possible applications in the emergency exhaust system. Based upon its high adsorption efficiency and retention properties, it is proposed that activated charcoal be utilized for iodine vapor adsorption.

- (p) Foam Suppression of Radioactive  
Iodine and Particulates  
R. Yoder, M. Fontana, L. Silverman  
NSA 18-16128

A method studied to remove radioactive halogens and particulates from air in reactor containment vessels is foam containment. In this method, an ether lauryl sulfate foam containing an iodine reactant is generated rapidly filling the entire volume. This provides a tremendous surface area to which the encapsulated gases and particles can diffuse and be removed. Tests were conducted in a 6000 cubic foot hot cell.





4-05 Miscellaneous Reactor Effluent Air Clean-up Systems

(a) A Multibed Low Velocity Air Cleaner

R. Yoder, F. Empson

Presented at Fifth A.E.C. Air Cleaning Conference

June 24-27, Boston, Mass.

NSA 12-12147

An air cleaner for the off-gases from processes which fix radioactive wastes in a sintered form for disposal is described. The components of the multibed cleaner are sand, soda lime, and activated carbon. Experimental data used in selecting the materials used are given and experiments with the cleaner are described. The device removes greater than 99.995% of particulates, and all fission product gases except krypton and xenon.

(b) Economic Survey of Air and Gas Cleaning

Operations Within A.E.C.

C. Billings, L. Silverman

Presented at Fifth A.E.C. Air Cleaning Conference

June 24-27, Boston, Mass.

NSA 12-12147

Plans for a survey of air and gas cleaning operations at AEC sites which is to be conducted is discussed. The information gathered will make it possible to establish design and cost criteria leading to economical, more efficient air and gas cleaning. A copy of a proposed questionnaire is included.

(c) Air Cleaning Costs - A Study of Three Systems

H. Jordan

Presented at Fifth A.E.C. Air Cleaning Conference

June 24-27, Boston, Mass.

NSA 12-12147

The air cleaning facilities at Los Alamos Scientific Laboratory for DP West Site, CMR Building, and Ten Site are described. The cost figures of each location are given including depreciation, operation, maintenance, and original cost of installation. The wet methods are found inherently more expensive than dry filtration.

(d) Experiments at ETR and MTR

D. Foster

Presented at Sixth A.E.C. Air Cleaning Conference

Boston, Mass. July 7-9, 1959

NSA 15-6245



The pre-stack disposal treatment of effluents from ETR and MTR tests of ANP fuel elements is described. It consists of a particle removal stage, iodine removal and a decay filter system. The filter system consists of two silver plated fiber flax beds with absolute filters and a delay tank between.

(e) Estimates of Accumulated Exposures and Environmental Build-up of Radioactivity

W. Culkowski

Presented at Sixth A.E.C. Air Cleaning Conference

Boston, Mass., July 7-9, 1959

NSA 15-6245

A simple method for estimating long-term effluent concentration and deposition values based on Sutton's and Chamberlin's equation is presented. Calculations of concentrations and deposition are straight forward, and become especially convenient if average wind data are employed.

(f) Argonne National Laboratory Air Cleaning Resume

C. Cheever

Presented at Sixth A.E.C. Air Cleaning Conference

Boston, Mass. July 7-9, 1959

NSA 15-6245

Various type of air cleaning equipment used at Argonne National Laboratory are described and air cleaning plans for Fuels Technology Center are discussed.

(g) Economic Survey of Air and Gas Cleaning

Operations Within A.E.C.

J. Fitzgerald, L. Silverman, R. Dennis, C. Billings

Presented at Sixth A.E.C. Air Cleaning Conference

Boston, Mass., July 7-9, 1959

NSA 15-6245

A number of AEC sites prepared analysis of the technical and economic aspects of their air cleaning activities. The data has been compiled and are to be summarized later. An appendix including some of this data is included.

(h) The Evaluation of Radioactive Releases

From Chemical Plants

E. Arnold, A. Gesky, J. Nichols

Presented at Seventh AEC Air Cleaning Conference

Brookhaven, Upton, N. Y. Oct. 10-12, 1961

T.I.D. 7627





A quantitative method for estimating the hazards associated with the maximum credible accident in a radio chemical facility was developed. The MCA in such facilities are chemical or nuclear explosions which disperse radioactive materials into ventilation systems. Approximate physical properties of these gases were combined with the efficiency of ventilation clean-up devices, and meteorological correlations to evaluate the hazards to the environment.

- (i) Fuel Element Decomposition Products  
G. Parker, G. Greek, W. Martin  
Presented at Seventh A.E.C. Air Cleaning Conference  
Brookhaven, Upton, N. Y. Oct. 10-12, 1961  
T.I.D. 7627

Reaction type fuels and their related hazards are discussed. Topics include product release from Uranium and Uranium Alloys.

- (j) The Containment of Fission Products  
In a Power Reactor  
S. Riley  
Presented at Seventh AEC Air Cleaning Conference  
Brookhaven, Upton, N. Y. Oct. 10-12, 1961  
T.I.D. 7627

Methods of removing iodine and other radioactive isotopes from gas streams in oxide fueled gas cooled reactors are discussed. The types of gas discharge that can occur under normal and emergency conditions are considered. Gas treatment processes are outlined.

- (k) The Analog Computer as an Aid  
In Critical Ventilation System Evaluation  
T. Pickel  
Presented at Eighth AEC Air Cleaning Conference  
Oak Ridge, Tennessee. Oct. 22-24, 1963  
T.I.D. 7677

Advantages of using analog computers in the analysis of air handling systems are discussed, and as an illustration of an analysis a simple system is presented.

- (l) Design Considerations for Exhaust Systems  
Involving Radioactive Particulates  
A. Fuller  
Presented at Eighth A.E.C. Air Cleaning Conference  
Oak Ridge, Tennessee October 22-24, 1963  
T.I.D. 7677



It is shown that to properly describe the efficiency of a filter in removing contaminants especially  $^{131}\text{I}$ , from an air stream not only must the amount of impurities be specified but also that the size distribution of particles in the air stream must be specified. Calculations and comparisons are presented.

- (m) Release of Fission Products on the In-Pile  
Melting of Reactor Fuels  
R. Shields, C. Miller, R. Lorenz, W. Browning  
Presented at Second Conference of Nuclear  
Reactor Chemistry, Gatlinburg, Tennessee, Oct. 10, 1961  
NSA 16-26472

Studies are being made to determine the fission product release characteristics of various fuels under conditions which simulate as closely as possible those which may exist at the time of a reactor incident. Fission product release when fuel materials are melted was investigated. The only experiment completed had a 3% enriched  $\text{UO}_2$  specimen. Examination of the test specimen indicated that a large part of the  $\text{UO}_2$  had undergone a change of state.

- (n) Emergency Actions in Radiation Accidents  
D. Davis  
As Included in Reactor Chemistry  
Division Annual Report Ending Jan. 21, 1964  
NSA 13726

Several definitions of a radiation emergency are given, and several improvements that should be made in emergency procedures are outlined. The AEC experience in radiation accidents and its methods for coping with them are described. Personnel dosimetry, exposures of rescue teams and radiation surveying of large areas are discussed.

- (o) Nuclear Merchant Ship Site Criteria  
W. Cottrell  
As Included in Reaction Chemistry  
Division Annual Report Ending Jan. 31, 1964  
NSA 13727

The site criteria for stationary reactors were modified in order to apply for the mobile reactor of NS Savannah in lieu of the city distance criterion. The three zones around the ship reactor are described. The conditions for port entry and additional measures for initial operations are outlined. The problems and implications of the criteria are discussed.



- p) Environmental Contamination Around Nuclear Facilities  
C. Barton  
As Included in Reactor Chemistry  
Division Annual Report Ending Jan. 31, 1964  
NSA 1373

The results of environmental surveys around nuclear facilities are described briefly. Stack monitoring data on the amounts of radioactive materials being discharged to the atmosphere are not readily obtainable, but a table of information on gaseous effluent monitoring and control practices at several reactor sites is presented. Contamination around fuel processing plants is considered briefly.

- q) The Present Status of Chemical Research in Atmospheric Purification and Control on Nuclear Powered Submarines  
V. Piatt, E. Ramskill  
NSA 16-33988

Research on submarine air-replenishment equipment is reported. Oxygen sources, carbon, dioxide removal, atmospheric sampling and analysis, and elimination of contaminants are discussed.

- r) Design and Test of a Gas Adsorption System  
for the N. S. Savannah  
G. Robinson  
Advances in Cryogenic Engineering 7(137-42)

This paper describes the design and engineering testing of a low temperature adsorption unit installed on the N. S. Savannah designed to remove and store fission-product gases for a 100-day period. Included is a description of the purification cycle, a discussion of design features and engineering test results, and a presentation of the design data for the adsorption of gases on activated charcoal of liquid nitrogen temperatures.

- s) Argon, Helium, and the Rare Gases Vol. 1.  
G. Cook - John Wiley & Sons (1961)

History, occurrence, and properties of the noble gases are discussed. Also included are solubility data and properties of the radio-isotops.



## SECTION V - SYSTEM DISCUSSION

### 5-01 General:

Five systems with a potential capability to perform the removal function in accordance with the data summarized in SECTION III - BASIS FOR DESIGN, were developed, reviewed, and evaluated. The systems studied, which are fully discussed in this Section, are summarized below:

#### System A - Low Temperature Solid Adsorption

This system depends on the direct adsorption of the radioactive noble gases on a refrigerated bed of solid adsorbent and is Not Recommended, since the adsorbent bed required is excessively large.

#### System B - Solid Adsorption with Cryogenic Enrichment

In this system, compact reliable cryogenic enrichment is used to concentrate the radioactive gases thereby reducing the adsorbent bed size requirements. This system is the Preferred Method, although a limited development program is required to obtain design data for the proper sizing of the adsorber beds.

#### System C - Cryogenic Distillation

The system for which the most operating experience and design information exists is the cryogenic distillation system. A system based on the principles of cryogenic distillation could probably be placed on stream in the shortest time; however, it is slightly more complex than System B and consequently would be more subject to mechanical failure. Furthermore, this system cannot be easily placed into immediate operation. Accordingly, this is the Second Choice System.

#### System D - Static Freon Adsorption System

In this system, the noble gases are removed from the contaminated air as it is bubbled through a liquid bath. Though this technique is quite effective on a laboratory scale, it is Not Recommended for this application.



### System E - Dynamic Freon Absorption System

A technically feasible system utilizes a counter-current Freon absorption tower. Though this system has several operational advantages, it can Not Be Recommended here, because there is not enough experimental data available at the present time to guide the detailed design and construction of a full scale system with any certainty of success.

### System F - Non-reversing Process Systems

In addition to the five systems discussed above, the utilization of non-reversing heat exchangers was also considered during the preliminary study stages, since certain advantages were apparent; namely, the use of non-reversing exchangers would guarantee containment of any radioactive iodine compounds or iodides which pass through the existing N. S. Savannah filtering system.

It is certain that these contaminants, present in minute quantities, would freeze out as the air is cooled to approximately  $-250^{\circ}\text{F}$ . If the exchangers are of the reversing type, there was an apparent danger that the iodides would be carried into the atmosphere during reversal. However, it has since been established that the iodides would probably plate out permanently on the exchanger surface. Therefore, the additional cost and complexity of a non-reversing system is not warranted.

For information purposes, non-reversing exchangers are shown as part of a cryogenic distillation system on Drawing No. 26 8821. It is equally possible to utilize non-reversing exchangers with any of the proposed systems.

### 5-02 System A - Low Temperature Solid Adsorbent System:

The simplest concept studied was the low temperature adsorption of the radioactive gases on a solid adsorbent; a minimum of moving parts and controls are required. A distinct advantage is that there are no fluids required that may result in sloshing due to the rocking motion of the ship.

In general, this system (shown on Drawing No. 26 8816) requires the compressing of the contaminated air to a pressure of about 35 psig and subsequent cooling in the precooler to a temperature of  $+40^{\circ}\text{F}$  to eliminate most of the moisture. Since the presence of carbon dioxide or water vapor adversely affects the capacity of the adsorbent bed, the air is cooled against the "cleaned-up,"





returning gas to approximately  $-290^{\circ}\text{F}$  in a reversing exchanger where the  $\text{CO}_2$  and water is deposited on the exchanger walls. To prevent excessive buildup of solids, the paths are periodically reversed and the  $\text{CO}_2$  and water deposited on the previous cycle is effectively removed by the clean returning gas. The cooled air is then passed through a bed of solid adsorbent (such as molecular sieves) where the noble gases are quantitatively adsorbed and contained as long as refrigeration is maintained. The effluent air, containing less than a tenth of a part per million noble gas, then passes through the exchanger system where it picks up the  $\text{CO}_2$  and water deposited in the previous cycle and cools the incoming air. The clean air is then exhausted to the atmosphere through the Savannah exhaust system. Liquid nitrogen from the shipboard storage tank provides the refrigeration for this system.

To contain the adsorbed noble gases for an indefinite period without liquid nitrogen, the adsorbent bed is warmed to approximately  $200^{\circ}\text{C}$  and the effluent gas compressed into a cylinder.

This system at first glance appears extremely simple and ideally suited to the Savannah. A detailed analysis, however, revealed several serious drawbacks which make this system unsuitable for this application. The most serious drawback of this system is the extremely large adsorbent bed required; the studies indicate that over 100 cubic feet of adsorbent material is required to reduce the noble gas concentration to a permissible level for a two-day operating period. The adsorption of noble gases from air is difficult since oxygen, and to a lesser extent, nitrogen - are also adsorbed on the surface. Consequently, the adsorption rate is limited by the relatively slow diffusion rate of the noble gases through the stagnant air film. This results in a long wave front and is aggravated by the extremely low concentration of the noble gases.

In addition, the large beds require substantial quantities of liquid nitrogen for refrigeration during cooldown and holding at approximately liquid oxygen temperatures. Therefore, the simple adsorption system is not recommended for this application.

#### System B - Cryogenic Enrichment Preceding Solid Adsorption:

As mentioned in the discussion of the previous system, solid adsorption has several desirable features but very large beds are required because of the relatively large gas flow and low noble gas concentration. In an effort to capitalize on the advantage of such a system, a simple cryogenic stripping column is used to enrich the noble gas concentration to about

80 ppm. This enrichment reduces the adsorber flow to about 26 scfm with a resultant reduction in the absorbent bed requirements to about 4 cubic feet.

This system is the Preferred System (see SECTION VIII - CONCLUSIONS AND RECOMMENDATIONS) and is described in detail in SECTION VI - DESIGN CRITERIA; for comparison with the other systems, it is briefly described here.

As shown on Drawing No. 26 8817, the contaminated air is compressed to about 30 psig and cooled to +40°F to remove most of the water vapor. The air then enters the reversing exchangers where the remaining water vapor and CO<sub>2</sub> are frozen out in the same manner as described for System A. The air is then filtered to remove particulates passed to the feed cooler, and cooled to -290°F. It then enters the bottom of a packed column. The contaminated air passes upwards counter-current to a descending stream of liquid nitrogen which condenses the relatively high boiling krypton and xenon and concentrates them in a relatively small oxygen-rich fraction in the bottom of the column. The gas leaving the top of the column (containing less than 0.10 of a part per million noble gases) passes through the exchanger system where it cools the inlet air and picks up the CO<sub>2</sub> and water deposited in the previous cycle.

The liquid from the bottom of the column is continuously withdrawn and vaporized. The resultant cold vapor is passed through a bed of molecular sieves where the noble gases are quantitatively adsorbed. The effluent air, free of noble gases, is exhausted from the unit after its refrigeration value is recovered while it chills the incoming air.

As in the previous system, the noble gases picked up during the two-day emergency will be firmly adsorbed on the surface of the absorbent as long as refrigeration is maintained. For permanent containment, the bed is heated to 200°C and the noble gases are driven off and compressed into cylinders by a small compressor. Drawing No. 26 8822 is a Piping and Instrumentation Diagram that describes this system in detail.

#### 5-04 System C - Cryogenic Distillation:

Krypton and xenon, though present in the air in only parts per million quantities, are separated and obtained in high yield commercially by cryogenic distillation. Air Reduction has built several such krypton producing units and is currently planning others. Consequently, considerable experience and data is available. As a result, it is clear that this system could be fabricated and installed in the shortest time; however, this system is more complex than the solid adsorption system with

cryogenic enrichment and would require more controls. Although this system would be subject to more mechanical malfunctions, it remains a sound "second choice."

As shown on Drawing No. 26 8818, effluent air from the containment room is compressed to 30 psig and then cooled in an aftercooler. The air is then further cooled in a precooler (to approximately  $+40^{\circ}\text{F}$  by cold air returning from the process) where most of the water is condensed and removed by a mechanical separator. The air is then deep-cooled to the reversing heat exchanger where the remaining water and carbon dioxide are frozen out (as described for the preceding processes).

A portion of this stream is returned to the precooler where it is warmed while cooling the inlet air. It then passes through a coil in the bottom of the distillation column to provide reboil before rejoining the main air stream.

The air is further cooled to about  $-290^{\circ}\text{F}$  in the preheating exchanger before it enters the distillation column. Liquid nitrogen from the shipboard liquid storage tank provides the refrigeration to condense about 15% of the vapors rising in the column to provide reflux. The refluxing action of the column washes the less volatile noble gases to the bottom of the column where they accumulate.

The uncondensed vapors are removed continuously from the top of the column and warmed slightly in the preheater to eliminate the formation of liquid in the reversing exchangers. The decontaminated air then passes through the reversing exchanger where it picks up the  $\text{CO}_2$  and water deposited in the previous cycle before being exhausted to the atmosphere.

The liquid in the bottom of the column, containing the accumulated noble gases, is periodically withdrawn and vaporized into cylinders capable of holding the gases at high pressures. The noble gases may be stored in this manner indefinitely.

Drawing No. 26 8823 is a Piping and Instrumentation Diagram depicting completely this system.

#### 05 System D - Static Freon Absorption System:

Since krypton and xenon are quite soluble in a number of solvents, the possibility of selectively dissolving the fission product noble gases was studied. Such a system would be very easy to start up and would result in a simple containment and disposal system. Though the noble gases are soluble in a host of solvents, the properties of Freon-12 were used for evaluation purposes.



The initial system considered utilized a simple batch-type absorber as shown on Drawing No. 26 8819. In this process, the contaminated air is compressed, cooled and then passed through a reversing heat exchanger to remove the water vapor. Since water is insoluble in Freon, it would be detrimental to the absorption process. Refrigeration for this operation is supplied from the shipboard liquid nitrogen storage tank.

The air is warmed to prevent freeze-up of the Freon bath in a second reversing heat exchanger by return stream air. The air then enters the bottom of the Freon filled absorber where the xenon and krypton are absorbed as the contaminated air bubbles up through the refrigerated Freon.

The effluent gas from the absorber enters the second reversing exchanger where any entrained Freon droplets are deposited. Upon cycle reversal, the deposited Freon is thawed and returned via Freon collectors to the main bath.

After leaving the second exchanger, the cleaned-up effluent gas passes through the first reversing exchanger where it picks up any CO<sub>2</sub> and water deposited in the previous cycle. It is then discharged to the atmosphere through the exhaust system of the ship.

The quantity of Freon in the absorber is sufficient to dissolve and contain the noble gas release for a full two-day emergency period. At the end of the operational period, the Freon could be transferred to a permanent storage vessel and stored until it is convenient to dispose of it.

It is evident that this system is quite adequate for relatively small flows, but uniformly distributing a flow of 200 scfm in a liquid is quite difficult without the use of a larger number of low capacity, high pressure drop spargers. This would result in a vessel with a very large cross-sectional area. In addition, since the contact time is directly proportional to the liquid depth, a large volume of Freon would be required. Consequently, this system is not considered suitable for this application.

#### -06 System E - Dynamic Freon Absorption System:

To retain the advantages of an absorption system, but reduce the quantity of Freon required, a dynamic system utilizing a packed column was developed. With this system, better contact between the liquid and gas is obtained.



As shown on Drawing No. 26 8820, the contaminated air is compressed, cooled in the precooler and passed through the reversing exchanger where the water vapor is removed. To prevent Freon freeze-up in the column, the cold air is then reheated in a second reversing exchanger.

The air then passes upwards counter-current to a descending stream of liquid Freon which quantitatively dissolves the noble gases. The Freon circulating system is designed to absorb and contain the entire quantity of noble gases released from the containment room during a full two-day emergency period.

The air which contains a small quantity of evaporated Freon, leaves the top of the absorber and enters the second reversing exchanger where it is cooled while depositing most of the entrained Freon on the exchanger walls. The Freon is then thawed during the next cycle by the incoming air and returned to the solvent system via the Freon collectors.

After leaving the second exchanger, the air passes through the reversing exchangers where it removes the water and any CO<sub>2</sub> deposited in the previous cycle. The air is then discharged to the atmosphere via the existing exhaust system.

Preliminary calculations indicate this process is definitely feasible and in general could result in a reliable, easy-to-operate system. It is estimated that a packed height of 15 feet in a 10-inch diameter column would give the required separation. This, of course, could be accomplished in several smaller columns if overall height was a limiting requirement. The Freon requirement is about 175 gallons circulated at a rate of 35 GPM.

This system, however, is not recommended for the N. S. Savannah since there is not enough data or experimental operating experience to justify the installation of such a unit at the present time.

#### 1-07 System F - Non-reversing Process Systems:

In all processes, the reversing heat exchangers may be replaced by conventional non-reversing exchangers if alternate means are provided for CO<sub>2</sub> and water removal. This generally is accomplished by the use of caustic scrubbers for CO<sub>2</sub> removal and industrial type dryers for moisture. The use of these higher cost systems are not warranted for this program.





## SECTION VI - DESIGN CRITERIA

### 6-01 General:

This section describes the criteria for the detailed design of a cryogenically enriched solid adsorption system for the removal and containment of noble gases from a 200 cfm gas stream.

Criteria is included for process development, built-in operational features, and equipment selection. The necessary design elements for safety and reliability are also included.

### 6-02 System Description: (See Drawing 26 8822) •

- (a) The process for the removal of the noble gases will depend on the adsorption of these gases by a bed of solid adsorbent after the noble gas concentration has been increased by cryogenic scrubbing in a packed column. The complete system will consist basically of a compressor, aftercooler, and receiver assembly, a precooler and water disposal system, reversing heat exchangers, a liquid nitrogen fed air scrubbing unit assembly, refrigerated adsorbent beds, a noble gas collection system, and a liquid nitrogen supply system.
- (b) The reversing heat exchangers, packed column assembly, and refrigerated adsorbent beds will be contained in an insulated cold box.
- (c) The compressor, aftercooler, receiver assembly will have sufficient capacity to remove the 200 cfm flow from the existing fan and filter assembly and to compress this flow to the required system operating pressure. A line open to the atmosphere through a spring loaded check valve will be provided to admit atmospheric air into the system if an excessively negative pressure exists due to a deficiency in the flow from the existing fan and filter system.
- (d) The precooler will have sufficient capacity to cool the air supplied by the compressor to 40°F. A water separator and disposal trap will be provided.
- (e) The reversing heat exchangers will have sufficient capacity to efficiently conserve refrigeration while removing CO<sub>2</sub> and water vapor by condensation and freezing on the heat transfer surface.

and, if necessary, by cold oxygen rich effluent from the adsorbent beds admitted through a control valve to an outer jacket in contact with the heat exchanger. A temperature controller will open this valve if additional cooling is required to maintain 40°F in the supply air stream or will open a control valve to bypass a portion of the nitrogen rich return stream if less cooling is required. The air leaving the precooler will flow through a water separator where entrained water will be removed and disposed of through a second liquid level type automatic drain trap.

- (c) The precooled and essentially dried air will pass through a reversing heat exchanger assembly where it will be cooled to close to the proper operating temperature of the scrubbing unit while depositing CO<sub>2</sub> and the balance of the water vapor on the surfaces of the exchanger. Refrigeration will be obtained from the nitrogen rich effluent gas from the wash tower. The heat exchanger passes will be periodically reversed by three-way valves under the control of a timer motor in order that the clean return gas may remove and exhaust the CO<sub>2</sub> and water vapor deposits. The deep cooled air will then pass through a small heat exchanger where it is further cooled by the oxygen rich liquid being withdrawn from the wash tower. The cold air will expand across a back pressure control valve and enter the wash tower where it will be scrubbed by liquid nitrogen supplied from the storage tank; the high boiling noble gases will be concentrated in an oxygen rich liquid at the bottom of the tower while clean nitrogen rich effluent gas will be withdrawn from the top and returned to the reversing heat exchanger assembly. The liquid level in the bottom of the wash tower will be controlled by a level controller that will open a liquid nitrogen supply valve to increase the level or open a valve in a line in parallel with the normal rich liquid withdrawal line to decrease the level.
- (d) The oxygen rich liquid containing the noble gases will be withdrawn from the tower, passed through the heat exchanger where it partially vaporizes while cooling the incoming air, and fully vaporized in a small exchanger heated by warm water discharged from the compressor cooling jackets. The cold vapor then will pass sequentially through a bank of adsorbent beds where the noble gases and some oxygen will be removed and retained by adsorption. The flow will be directed through one adsorbent tube after the other, through valves under the control of a timer motor. The oxygen rich effluent gas will pass through the precooler temperature control valve if required for refrigeration or directly to the exhaust system. A small amount of liquid nitrogen will be admitted through a control valve, vaporized in the space surrounding the adsorbent containers, and vented into the exhaust system.



- (f) The air scrubbing unit assembly will consist of a wash tower with a feed nitrogen control valve, a feed air refrigeration coil assembly, a feed air back pressure control valve, a liquid level control system, and a rich oxygen withdrawal system and vaporizer.
- (g) The solid adsorbent beds will consist of multiple tubes packed with a suitable adsorbent material and contained within a pressure tight casing.
- (h) The noble gas collection system will consist of a compressor, aftercooler, and receiver (storage tank). The compressor will have sufficient capacity to remove the gases released during the warming of the adsorbent beds in a reasonable time. The receiver will be sized to contain the gases released at the discharge pressure of the compressor.
- (i) The liquid nitrogen supply system will consist of a double walled storage vessel, a pressure building station, and a vacuum pump to maintain a vacuum in the insulation space between the double walls. The storage vessel will be sized to contain the liquid nitrogen required for a two-day operation of the system with a reserve for losses due to heat leak during a 20-day period preceding operation. The pressure building station will have sufficient capacity to maintain a pressure of 60 psig in the storage tank while supplying sufficient warm nitrogen gas to operate the instrumentation system and control valves.

6-03 System Operation:

- (a) Prior to an accident, the exhaust gas from the existing fan and filter assembly will be bypassed around the system and discharged directly to the N. S. Savannah exhaust stack. The system will be de-energized and maintained in a ready to operate condition. Vent gas from the liquid nitrogen storage tank will circulate through the components in the cold box to keep them as cool as possible.
- b) Upon actuation of the system start button, the main circulation compressor will be started and the compressor discharge shut-off valve will open. The compressed air will flow through an after-cooler where the compressor heat of compression will be removed and a certain amount of moisture will be separated out and disposed of through a liquid level type automatic drain trap. The gas will then pass through a receiver to a pre-cooler where it will be cooled to approximately +40°F by nitrogen rich air returning from the wash tower via the reversing heat exchangers



and, if necessary, by cold oxygen rich effluent from the adsorbent beds admitted through a control valve to an outer jacket in contact with the heat exchanger. A temperature controller will open this valve if additional cooling is required to maintain 40°F in the supply air stream or will open a control valve to bypass a portion of the nitrogen rich return stream if less cooling is required. The air leaving the precooler will flow through a water separator where entrained water will be removed and disposed of through a second liquid level type automatic drain trap.

- (c) The precooled and essentially dried air will pass through a reversing heat exchanger assembly where it will be cooled to close to the proper operating temperature of the scrubbing unit while depositing CO<sub>2</sub> and the balance of the water vapor on the surfaces of the exchanger. Refrigeration will be obtained from the nitrogen rich effluent gas from the wash tower. The heat exchanger passes will be periodically reversed by three-way valves under the control of a timer motor in order that the clean return gas may remove and exhaust the CO<sub>2</sub> and water vapor deposits. The deep cooled air will then pass through a small heat exchanger where it is further cooled by the oxygen rich liquid being withdrawn from the wash tower. The cold air will expand across a back pressure control valve and enter the wash tower where it will be scrubbed by liquid nitrogen supplied from the storage tank; the high boiling noble gases will be concentrated in an oxygen rich liquid at the bottom of the tower while clean nitrogen rich effluent gas will be withdrawn from the top and returned to the reversing heat exchanger assembly. The liquid level in the bottom of the wash tower will be controlled by a level controller that will open a liquid nitrogen supply valve to increase the level or open a valve in a line in parallel with the normal rich liquid withdrawal line to decrease the level.
- (d) The oxygen rich liquid containing the noble gases will be withdrawn from the tower, passed through the heat exchanger where it partially vaporizes while cooling the incoming air, and fully vaporized in a small exchanger heated by warm water discharged from the compressor cooling jackets. The cold vapor then will pass sequentially through a bank of adsorbent beds where the noble gases and some oxygen will be removed and retained by adsorption. The flow will be directed through one adsorbent tube after the other, through valves under the control of a timer motor. The oxygen rich effluent gas will pass through the precooler temperature control valve if required for refrigeration or directly to the exhaust system. A small amount of liquid nitrogen will be admitted through a control valve, vaporized in the space surrounding the adsorbent containers, and vented into the exhaust system.



- (e) At the conclusion of the removal period, shut-off valves will close to isolate the contaminated adsorbent beds, the compressor discharge shut-off valve will close, and warm compressor air will pass through a shut-off valve to the adsorbent bed casing to heat the beds and then through a second shut-off valve to the exhaust system. The contaminated gases released from the adsorbent material will be carried by a small flow of purge nitrogen gas and drawn through a shut-off valve to the collection system compressor. The gases will be compressed, aftercooled, and stored in high pressure bottles.
- (f) At the conclusion of the purge cycle, the system will be returned to a standby condition. The contaminated gases may be disposed of at any safe time.

#### 6-04 Main Compressor Assembly:

- (a) The main compressor will be a Joy Model WCOL-9 or equal vertical, single-stage, non-lubricated, water cooled compressor with an actual capacity of 200 cfm at 14.5 psia inlet and 45 psia discharge. The compressor will be driven by a 25 horsepower, 1800 RPM, motor.
- (b) The compressor will be furnished complete with V-belt drive, belt guard, aftercooler with moisture separator and automatic condensate trap, ASME coded receiver, discharge relief valve, automatic unloading system with pressure switch, discharge pressure gauge, high discharge air temperature switches, high cooling water temperature switch, low discharge pressure switch, vibrator, pulsation bottles, and cooling water temperature control valve.

#### 6-05 Cold Box Assembly:

- (a) The reversing heat exchangers, the wash tower assembly, and the adsorbent beds will be installed within an insulated cold box constructed of sheet metal and sealed against moisture penetration. A small nitrogen purge connection will be provided to eliminate residual vapors. The cold box assembly will be structurally self-supporting, provided with access panels to facilitate maintenance, and provided with insulation fill and drain connections. All valve operators (except solenoids) will be mounted external to the cold box. The precooler assembly and associated control valves and piping will be mounted on the side of the cold box.





- (b) The precooler assembly will consist of a Joy-Collins or equal three pass heat exchanger, a water separator, and an automatic liquid level type drain system. The basic precooler coil will be capable of cooling 200 scfm of air saturated at 95°F and approximately 40 psia to 40°F while warming 206 scfm of air supplied saturated at +25°F and approximately 20 psia. The total pressure drop of both streams will not exceed 6 psi. The third exchanger pass shall provide intimate contact between 26 cfm of oxygen rich gas at approximately -300°F and the warmer supply air in such a manner that the supply air may be cooled from +50°F to +40°F if a deficiency in temperature or flow exists in the primary refrigeration stream. The pressure drop in this auxiliary pass will not exceed 1 psi. The water separator will be capable of removing entrained water from the 200 scfm precooled air stream and delivering it to the automatic drain assembly. This assembly will be capable of discharging the collected water to an atmospheric drain system at the rates required for continuous operation without substantial pressure fluctuation in the piping system.
- (c) The reversing heat exchangers will be compact Trane or equal aluminum core with flanged connections. The exchangers will be capable of cooling 200 scfm of CO<sub>2</sub> and water vapor contaminated air supplied at approximately 40 psia from +40°F to -293°F while warming 206 scfm of an 85 per cent nitrogen, 15% oxygen mixture supplied at approximately 25 psia from -297°F to +25°F. The total pressure drop in both streams will not exceed 10 psi. The exchanger surface will be arranged to provide a reasonable run time between pass reversals.
- (d) The air scrubbing unit assembly will be capable of concentrating the noble gases in a 200 scfm contaminated air stream into an oxygen rich liquid when provided with approximately 2 lbs. per minute of liquid nitrogen for reflux. The assembly will be further capable of vaporizing approximately 2 lbs. per minute of the rich liquid for subsequent supply to the adsorbent beds. The assembly will be constructed of stainless steel and rated for a minimum operating pressure of 40 psig. The column will be of the packed type and provided with a full diameter access flange. The vaporizer will be capable of vaporizing the specified flow of liquid when provided with warm water discharged from the compressor cooling jackets. An electrical heater and control will be provided to guard against freezing.
- (e) The adsorbent beds will be capable of adsorbing and firmly containing the noble gases in a 20 scfm oxygen-nitrogen mixture supplied at a temperature of approximately -290°F continuously for 48 hours. The adsorbent material will be contained within



copper tubes provided with suitable screens that are in turn housed within a pressure tight casing. The casing will be designed to withstand an internal pressure of 5 psig and will be provided with a liquid nitrogen fill line and shut-off valve, a warm air inlet line and shut-off valve, a drain line and shut-off valve, and a vent line.

6-06 Collection System Assembly:

- (a) The collection system compressor will be a Corblin Model AICV 250 or equal two-stage diaphragm-type compressor with an actual capacity of 1 cfm at 14.7 psia inlet and 2400 psia discharge. The compressor will be driven by a 3 horsepower, 1800 RPM, motor.
- (b) The compressor will be furnished complete with V-belt drive, belt guard, aftercooler, interstage and discharge relief valves, interstage and discharge pressure gauges, automatic unloading system, and high and low discharge pressure switches.
- (c) The high pressure gas storage bottles will be capable of containing approximately 1000 scf of compressed gas at 2200 psi indefinitely. The heat generated due to the radioactive decay of the fission products will be dispersed into the surroundings by natural convection augmented if required by a thermostatically controlled blower or exhaust hood arrangement.

6-07 Liquid Nitrogen Supply System:

- (a) The liquid nitrogen storage vessel will have a minimum capacity of 1250 gallons exclusive of a 10% ullage space. The inner vessel will be constructed of stainless steel and ASME code stamped for operation at 60 psig minimum. The vessel will be provided with liquid fill and withdrawal connections, back pressure control valve, pressure relief valve, rupture disc, and vaporizer connections. The losses due to heat leak will not exceed .75% of full capacity per day.
- (b) The vaporizer will be of adequate size to provide approximately 200 scfh of normal temperature gas on a continuous basis while receiving heat from the surrounding air.



6-08 Instrumentation and Controls:

- (a) The instruments necessary for the control of the system with the exception of certain local temperature and pressure switches will be mounted on a free standing control panel. This panel will also contain the pushbuttons required to start the system as well as the system status indicating lights.
- (b) All pertinent temperatures will be monitored by a Minneapolis-Honeywell or equal 24 point strip chart recorder.
- (c) The back pressure control valves in the line entering the scrubbing unit and the vent line from the liquid nitrogen storage tank will be positioned by Fisher-Wizard or equal pressure controllers.
- (d) The valves regulating the liquid level in the scrubbing unit will be positioned by a Barton Controls or equal differential pressure controller.
- (e) The valves controlling the temperature of the air leaving the pre-cooler and the temperature of the oxygen rich gas leaving the vaporizer will be positioned by Minneapolis-Honeywell or equal temperature controllers.
- (f) A West or equal pyrometer will cycle a solenoid to admit liquid nitrogen into the casing surrounding the adsorbent beds upon demand.
- (g) All pertinent system pressures will be indicated on 4 1/2 inch dial gauges.

6-09 Gas Chromatograph:

- (a) The effluent gas stream will be monitored by an Air Reduction Model 22-7701 Gas Chromatograph.
- (b) The chromatograph will have a scale deflection for krypton of 24 divisions or greater for krypton in standard air (1.13 ppm) and will be repeatable to  $\pm 2$  divisions or better. Therefore, it will detect traces of krypton in air of 0.1 ppm or less (0.05 with operator care).



-10 Piping:

- ) The piping systems will be designed and tested for at least 100 per cent higher than the maximum system operating pressure.
- ) All warm piping will be carbon steel or copper. All cold piping will be stainless steel, aluminum, or copper.
- ) Relief valves will be provided in the piping systems to guard against excessive pressures that can result due to valve isolation.

11 Electrical:

- ) All electrical work will be in accordance with the National Electrical Code and any other codes governing shipboard use.
- ) The switchgear (motor starters) controlling the compressor motors will be mounted on the compressor skids. All relays and controls necessary for the operation of the system will be mounted in a standard NEMA enclosure installed within the control panel.

12 Testing:

- ) The system will be completely assembled and tested prior to delivery for shipboard installation.
- ) The compressors will be operated and checked for capacity against the specified delivery pressure.
- ) The cold box assembly will be proof pressure tested and leak tested.

The assembly will be operated with standard air (1.13 ppm krypton), all controls will be adjusted, and the effluent gas stream will be monitored for krypton content.

After the unit is functioning properly, a mixture of approximately 1% krypton in nitrogen will be introduced into the 200 cfm air stream to increase the krypton concentration to about 10 ppm. The effluent gas stream will be monitored for krypton content for a period of 2 days.



- (f) After the 2-day operating period, the purge and collection system will be operated to desorb the krypton from the absorbent beds. The purge gas will be monitored to demonstrate that clean up is complete.





## SECTION VII - COST ANALYSIS AND SCHEDULE

### 7-01 General:

Cost analyses were prepared for the two most attractive systems (System B and System C) previously described in SECTION V - SYSTEMS DISCUSSION of this report. These cost analyses were in turn a basis for the final system selection as described in SECTION VI - DESIGN CRITERIA. Presented herein are the anticipated costs to be incurred for the detailed design, fabrication and allowances for the pre-installation tests, and final installation and check-out aboard the N. S. Savannah ready for operational service.

An additional cost analysis was made which indicates the savings which could be accomplished should the basic 200 scfm purge flow rate be reduced to 100 scfm or less (see 7-04).

A schedule of operations is presented. This schedule shows the time required to complete the design, fabrication, installation and test of the described unit.

### 7-02 System B - Solid Adsorption with Cryogenic Enrichment:

Detail Design \$ 16,000

#### Material

a. LN <sub>2</sub> Tank, Vacuum Jacketed Piping, Pumps and Auxiliary Equipment	\$ 16,000
b. Main Compressor Equipment	9,600
c. Instrumentation and Control Valves	12,200
d. Cold Box, Exchangers, Adsorbent Towers, etc.	9,600
e. Collection Systems	8,200
f. Piping and Electrical	2,900
g. Analytical Equipment	6,000
	\$ 64,500

Fabrication Labor	8,900
Test	7,000
Travel and Subsistence	1,700
Allowance for Overhead & Profit	23,000
Shipboard Installation Cost	6,500

Estimated Project Cost \$ 127,600



7-03 System C - Cryogenic Distillation:

Detail Design		\$ 17,500
Material		
a. LN <sub>2</sub> Tank, Vacuum Jacketed Piping Pumps and Auxiliary Equipment	\$ 16,000	
b. Main Compressor Equipment	9,600	
c. Instrumentation and Control Valves	14,000	
d. Cold Box, Exchangers Column, etc.	11,400	
e. Collection System	7,500	
f. Piping and Electrical	3,100	
g. Analytical Equipment	6,000	
		67,600
Fabrication Labor		14,200
Test		12,800
Travel and Subsistence		1,900
Allowance for Overhead & Profit		29,000
Shipboard Installation Cost		7,500
Estimated Project Cost		\$ 150,500

7-04 100 SCFM Solid Adsorption System with Cryogenic Enrichment:

(Alternate Based on System B)

Detail Design		\$ 16,000
Materials		
a. LN <sub>2</sub> Tank, Vacuum Jacketed Piping, Pumps and Auxiliary Equipment	\$ 13,000	
b. Main Compressor Equipment	7,000	
c. Instrumentation and Control Valves	11,000	
d. Cold Box, Exchangers, Adsorbent Towers, Etc.	7,000	
e. Collection System	7,200	
f. Piping and Electrical	2,000	
g. Analytical Equipment	6,000	
		\$ 53,200
Fabrication Labor		7,000
Test		5,500
Travel and Subsistence		1,700
Allowance for Overhead and Profit		19,500
Shipboard Installation Cost		6,000
Estimated Project Cost		\$ 108,900



7-05 Schedule of Operations:

A detailed analysis of the design, fabrication, test, and installation requirements has resulted in the following realistic schedule:

<u>Operation</u>	<u>Elapsed Time</u> <u>(Weeks)</u>
Initiate Design	0
Process Review Completed	3
Detailed Design Completed	9
Initiate Procurement	3
Initiate Fabrication	6
Fabrication Completed	12
Inspection Completed	13
Assembly for Test Completed	15
Pre-delivery Testing Completed	17
Initiate Installation of Supporting Services	16
Delivery	18
System Installation Completed	20
Shipboard Tests Completed	21
Personnel Training	21



## SECTION VIII - CONCLUSIONS

The study for determining the feasibility of recovering the noble gases, evolved during a malfunction of the nuclear reactor on the N. S. Savannah, has been completed. Several systems with varying degrees of reliability and shipboard adaptability have been investigated. Some of these systems were eliminated from final consideration due to various deficiencies in reliability or adaptability for their intended use. Two of the investigated systems closely approach the criteria as presented. A final evaluation based on reliability, ease of operation and maintenance, and initial cost resulted in the selection of System B.- "Solid Adsorption With Cryogenic Enrichment."

This system has a high degree of reliability and will meet the needs of the N. S. Savannah with a minimum of development effort. The reversing exchangers for CO<sub>2</sub> and water removal are widely used in the air separation industry and present no design or operating problem for this application. Liquid nitrogen wash columns have successfully been used in the chemical industry and as a result considerable design data is available.

The absorbent beds cannot be precisely sized by analysis with a high degree of certainty; therefore, pre-installation factory tests of the unit will be required. During the testing period, the beds will be checked for efficiency of removal of noble gases, for total absorption capacity, for pressure drop, and for clean-up ability. Some modification of the beds may be required.

This program can be conducted concurrently with the on-board installation of supporting services and equipment and will not seriously affect the overall design, fabrication, and installation program.



SECTION IX - DRAWINGS

FLOW DIAGRAMS

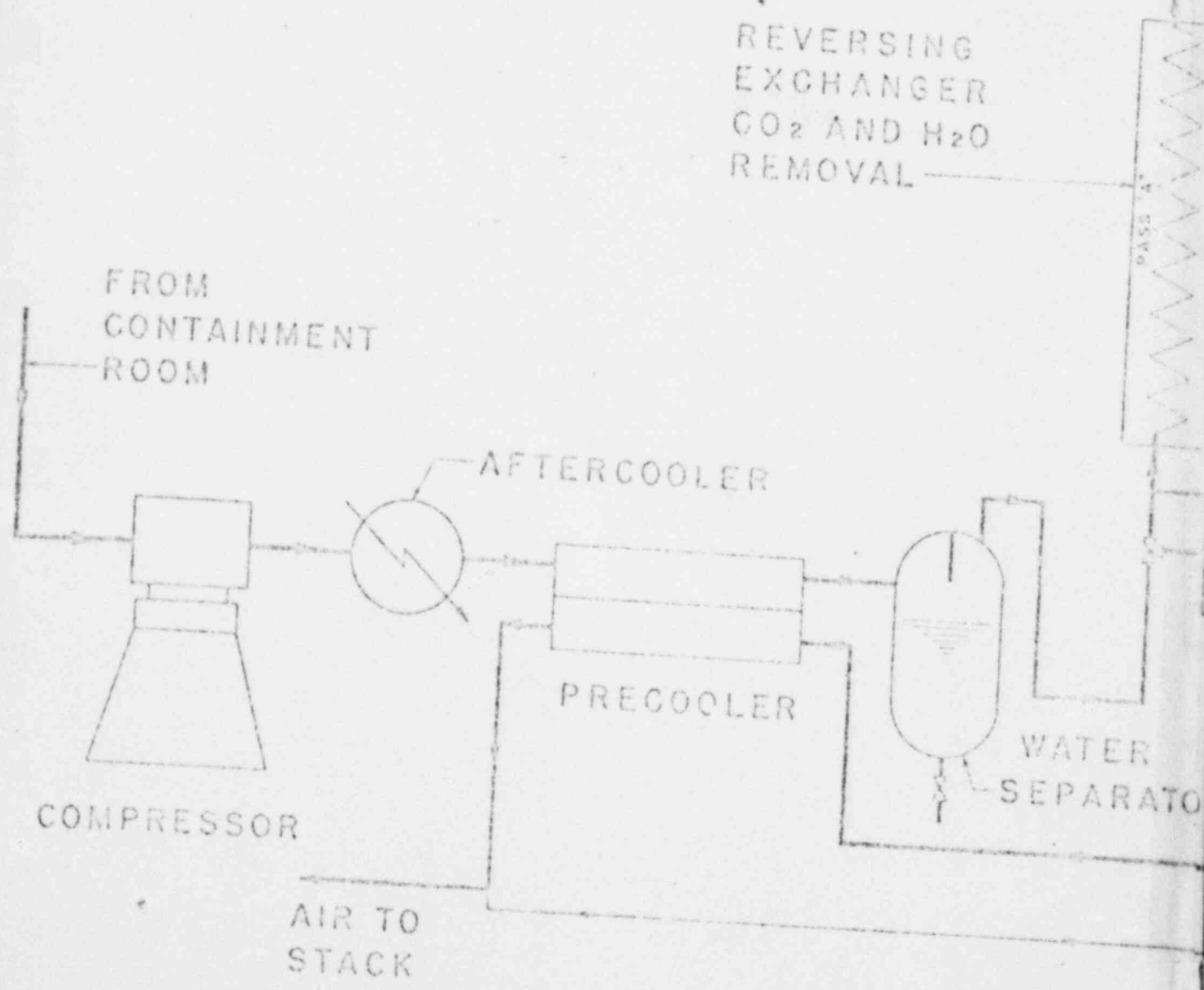
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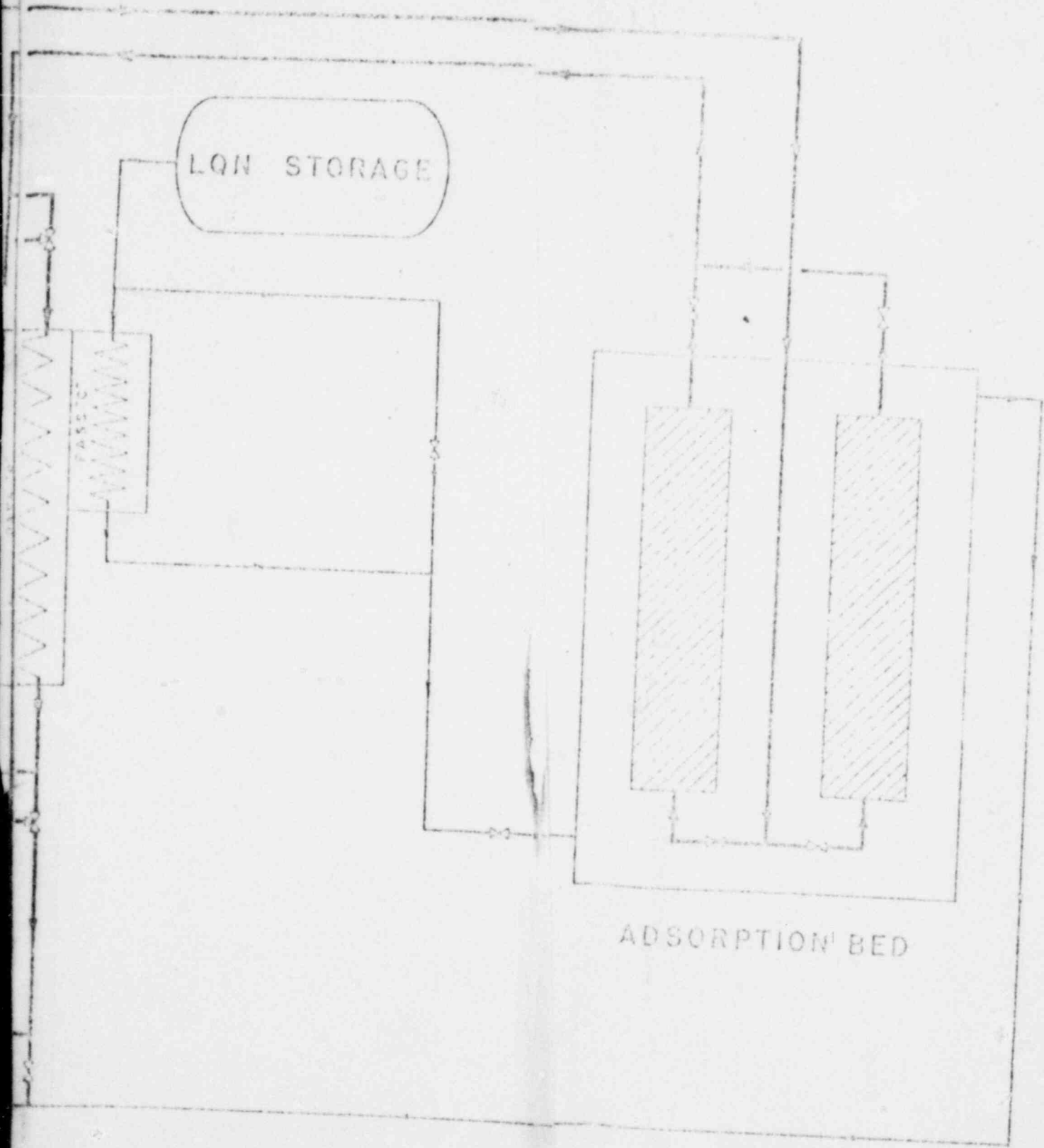
System A - Low Temperature Solid Adsorption	D 26-8816
System B - Solid Adsorption with Cryogenic Enrichment	D 26-8817
System C - Cryogenic Distillation	D 26-8818
System D - Static Freon Absorption	D 26-8819
System E - Dynamic Freon Absorption	D 26-8820
System F - Non-Reversing Process	D 26-8821

PIPING AND INSTRUMENT DIAGRAMS

System B - Solid Adsorption with Cryogenic Enrichment	D 26-8822
System C - Cryogenic Distillation	D 26-8823



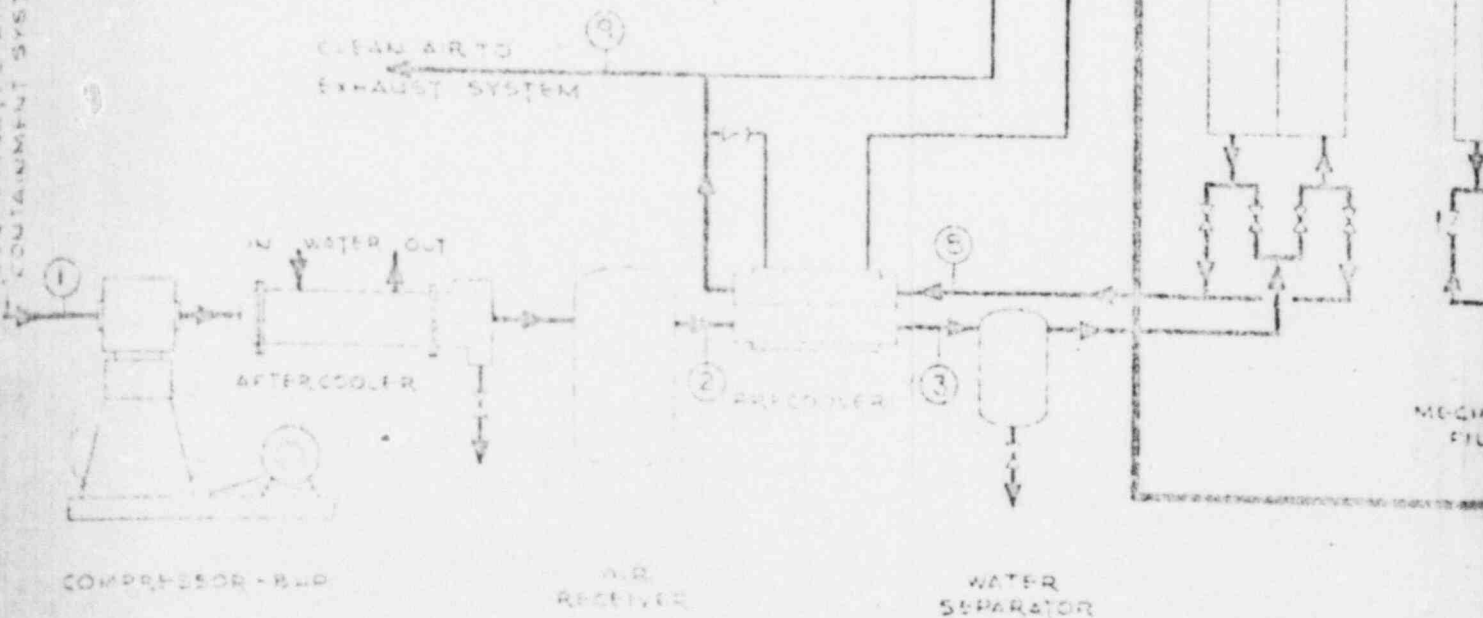


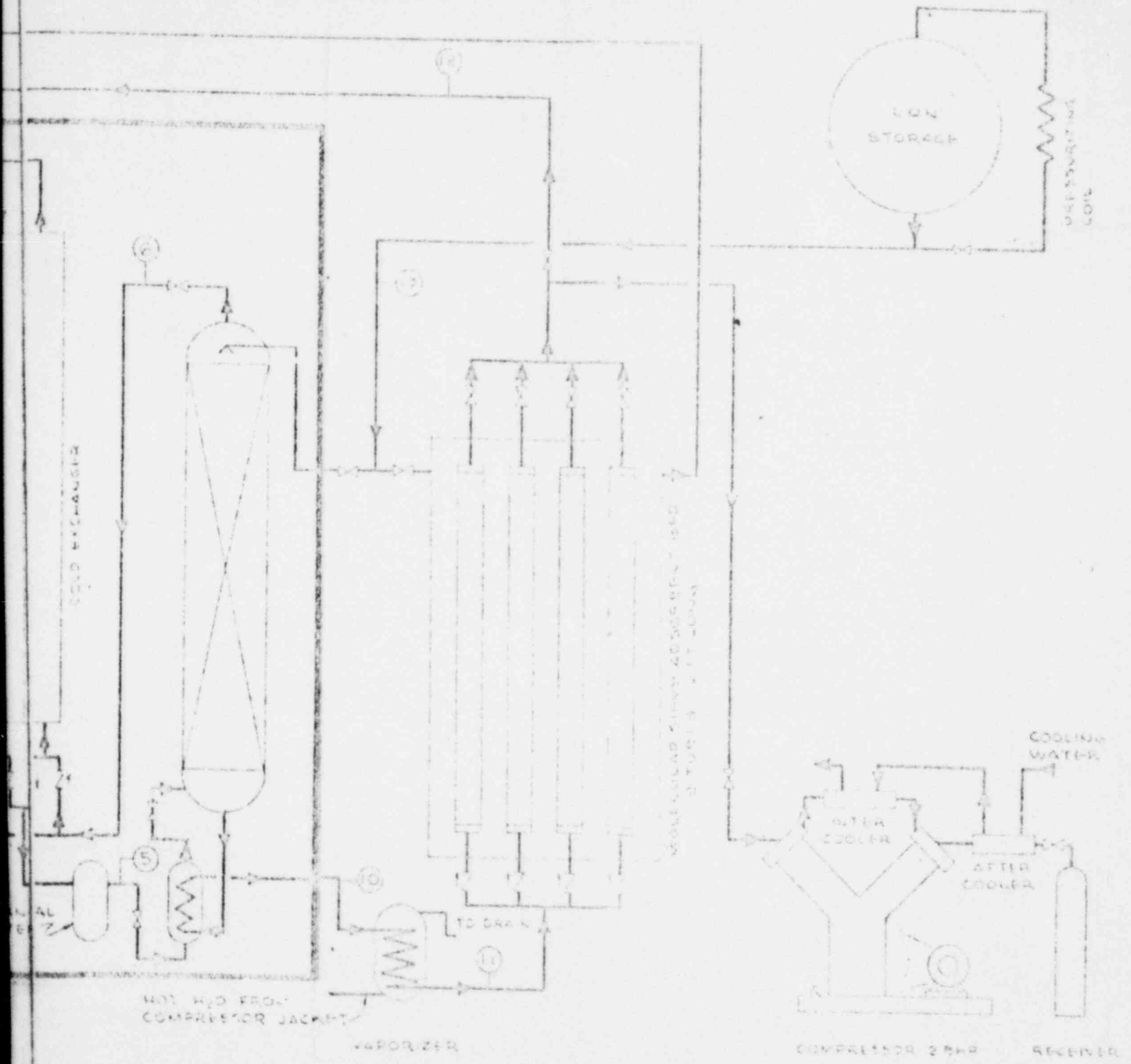
M. S. SAVANNAH  
 NOBLE GAS RECOVERY  
 SYSTEM A  
 LOW TEMPERATURE  
 SOLID ADSORPTION

LINE NO	DESIGNATION	TEMPERATURE (°C)	PRESSURE (PSIA)
1	AIR FROM CONTAINMENT ROOM	100	147
2	DISCHARGE FROM COMPRESSOR	200	40
3	AIR TO COLD BOX	100	27
4	EXCHANGER MID POINT	100	28
5	AIR TO SCRUBBING COLUMN	100	27
6	CLEAN AIR TO COLD PRE HEATER	100	27
7	EXCHANGER MID POINT	100	21
8	COLD AIR TO PRECOOLER	25	16
9	CLEAN AIR TO AIR EXHAUST	50	14
10	CONCENTRATED LIQUID STREAM	25	25
11	VAPORIZER STREAM TO ADSORBER	100	24
12	ADSORBER EFFLUENT GAS	100	10
13	LIQUID NITROGEN REFLUX	100	50

CONTAMINATED AIR FROM CONTAINMENT SYSTEM



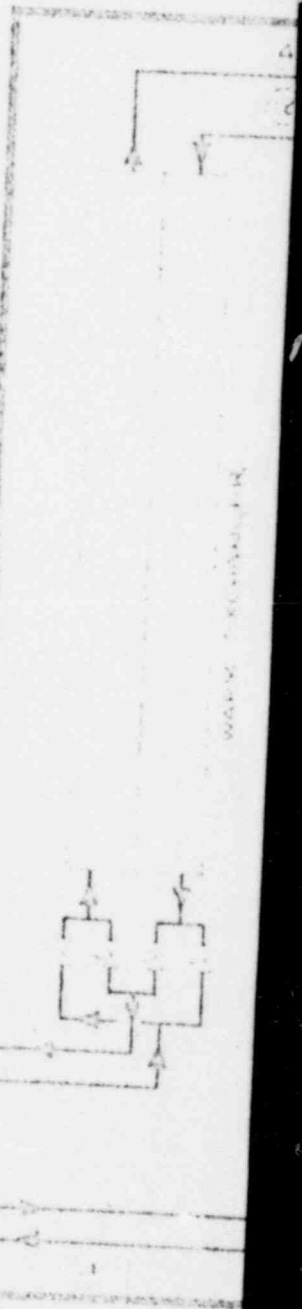
NO.	DATE	BY	REVISION



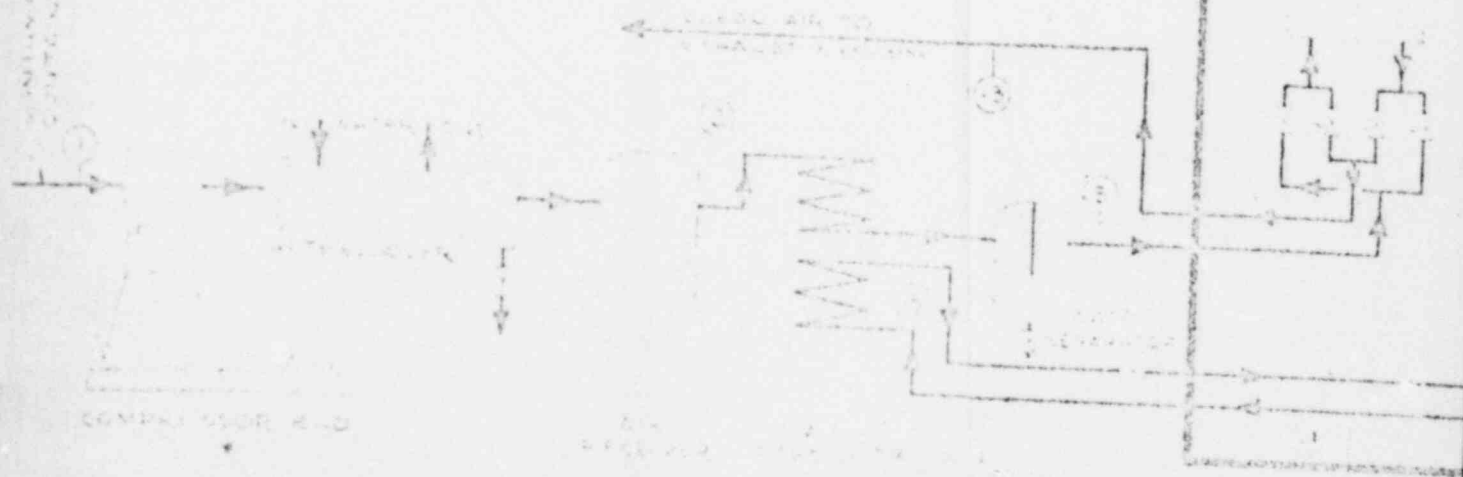
NO. LEVINGDON  
 NOBLE GAS RECOVERY  
**SYSTEM B**  
 SOLID ADSORPTION WITH  
 CRYOGENIC ENRICHMENT  
 RECEIVED FROM SYSTEM A  
 A.S. RESEARCH SALES COMPANY

COMPONENT	NUMBER	DESCRIPTION	UNIT

LINE NO.	DESCRIPTION	UNIT
15	EXHAUST FLOW	1000 CFM
1	AIR FROM CONTAINMENT ROOM	1000 CFM
2	DISCHARGE FROM COMPRESSOR	1000 CFM
3	AIR TO COLD ROOM	400 CFM
4	PREHEATED AIR FLOW	175 CFM
5	PREHEATED AIR FLOW	175 CFM
6	AIR TO WASH VALVE	1000 CFM
7	AIR TO HEAT EXCHANGER COLUMN	1000 CFM
8	COLUMN AIR FLOW	1000 CFM
9	EXHAUSTER DISCHARGE	1000 CFM
10	CONTAINMENT SYSTEM EXHAUST	1000 CFM
11	WARM AIR TO COLD EXCHANGER	1000 CFM
12	WARM AIR TO FLOW	1000 CFM
13	WARM AIR TO COLD EXHAUST	1000 CFM
14	COLD AIR TO PREHEATER	1000 CFM
15	WARM AIR TO WASHOVER	1000 CFM
16	WARM AIR TO WASH VALVE	1000 CFM
17	AIR TO EXHAUSTER	1000 CFM
18	COLD AIR TO COLD EXCHANGER	1000 CFM
19	COLD AIR TO COLD EXHAUST	1000 CFM
20	AIR FLOW TO EXHAUST	1000 CFM



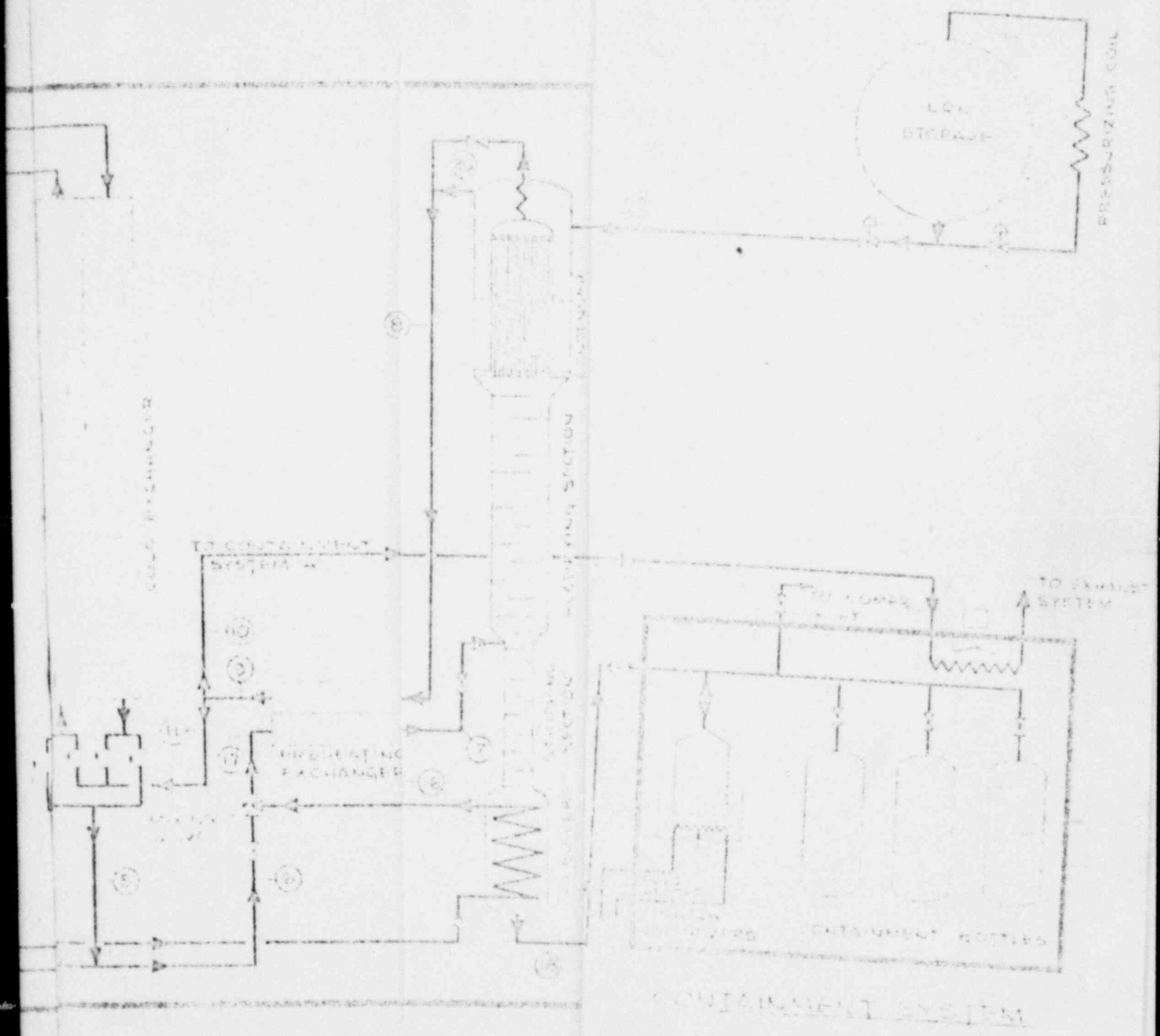
CONTAINMENT ROOM AIR FLOW



CONTAINMENT ROOM AIR FLOW

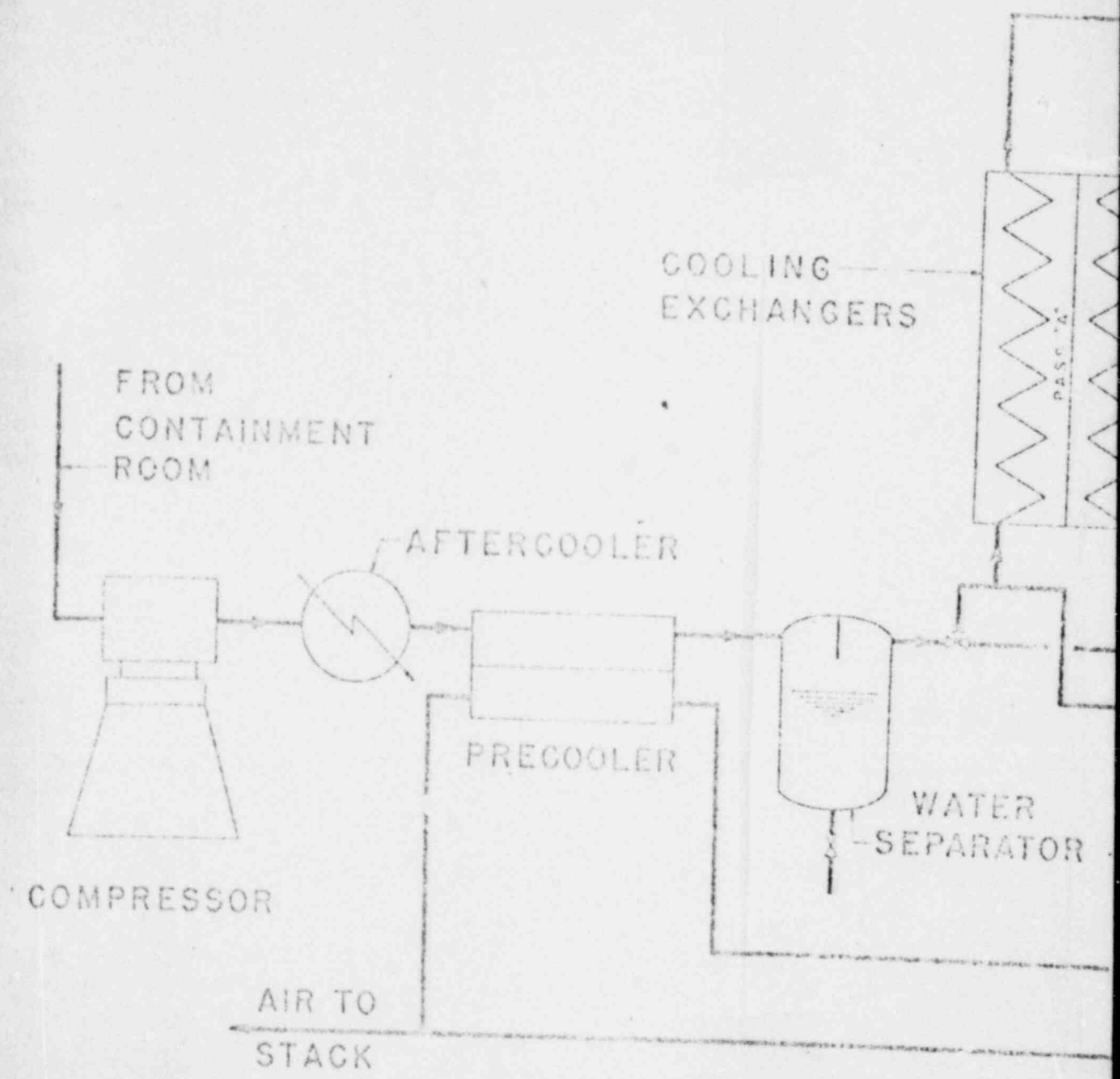


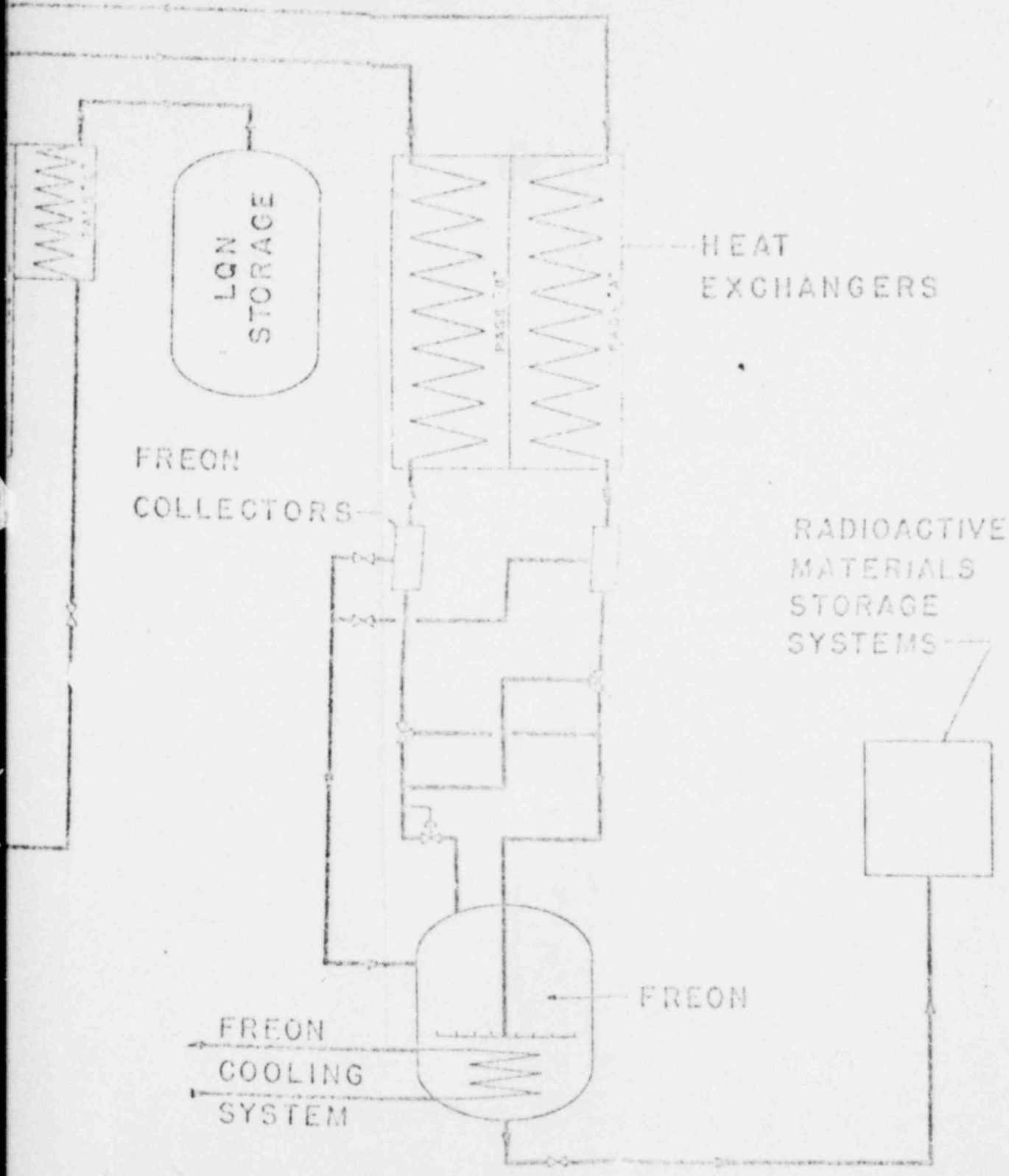
NO. 100-1000		
DATE		
REVISION		
BY		
CHECKED		



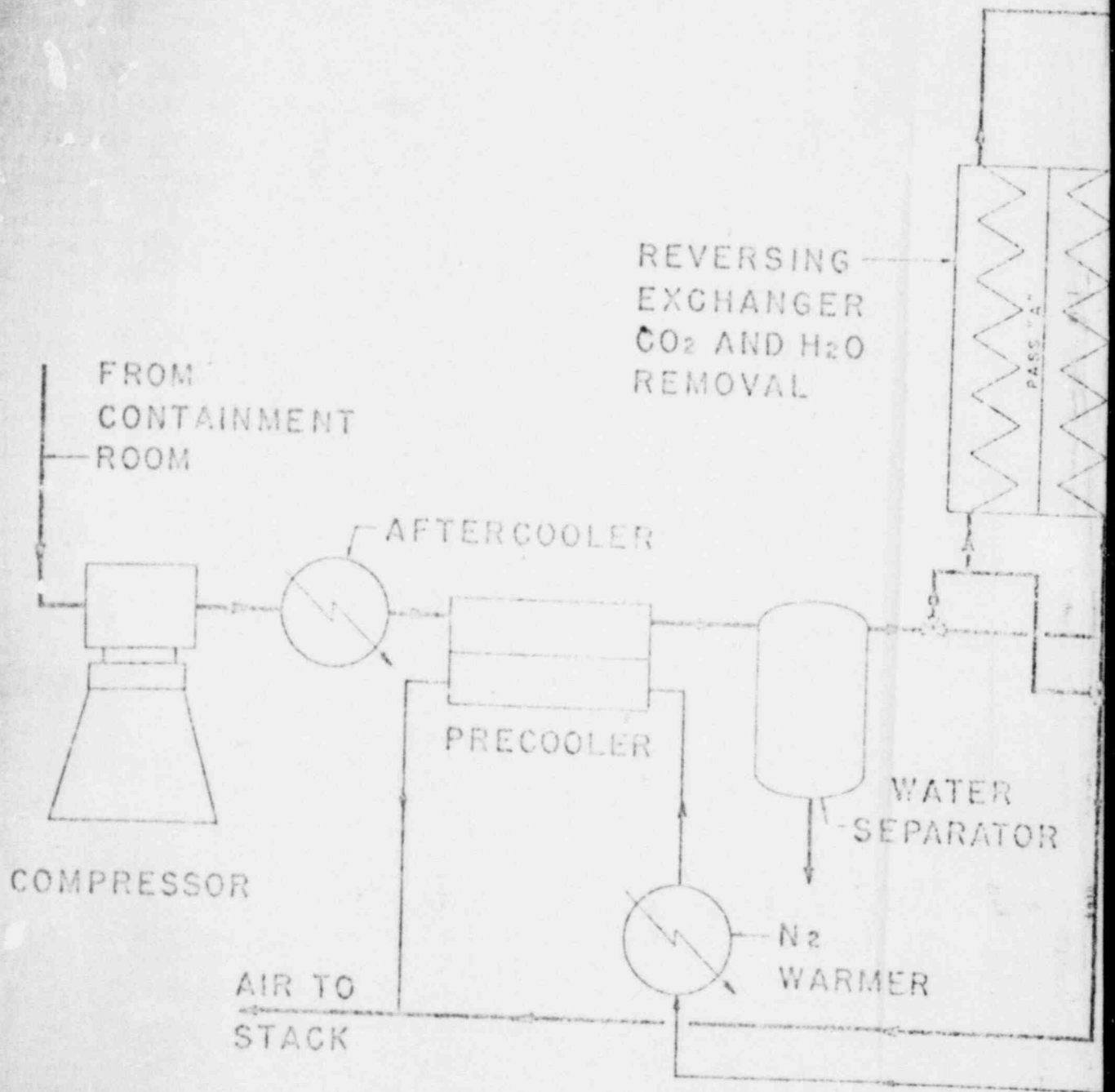
CONTAINMENT SYSTEM

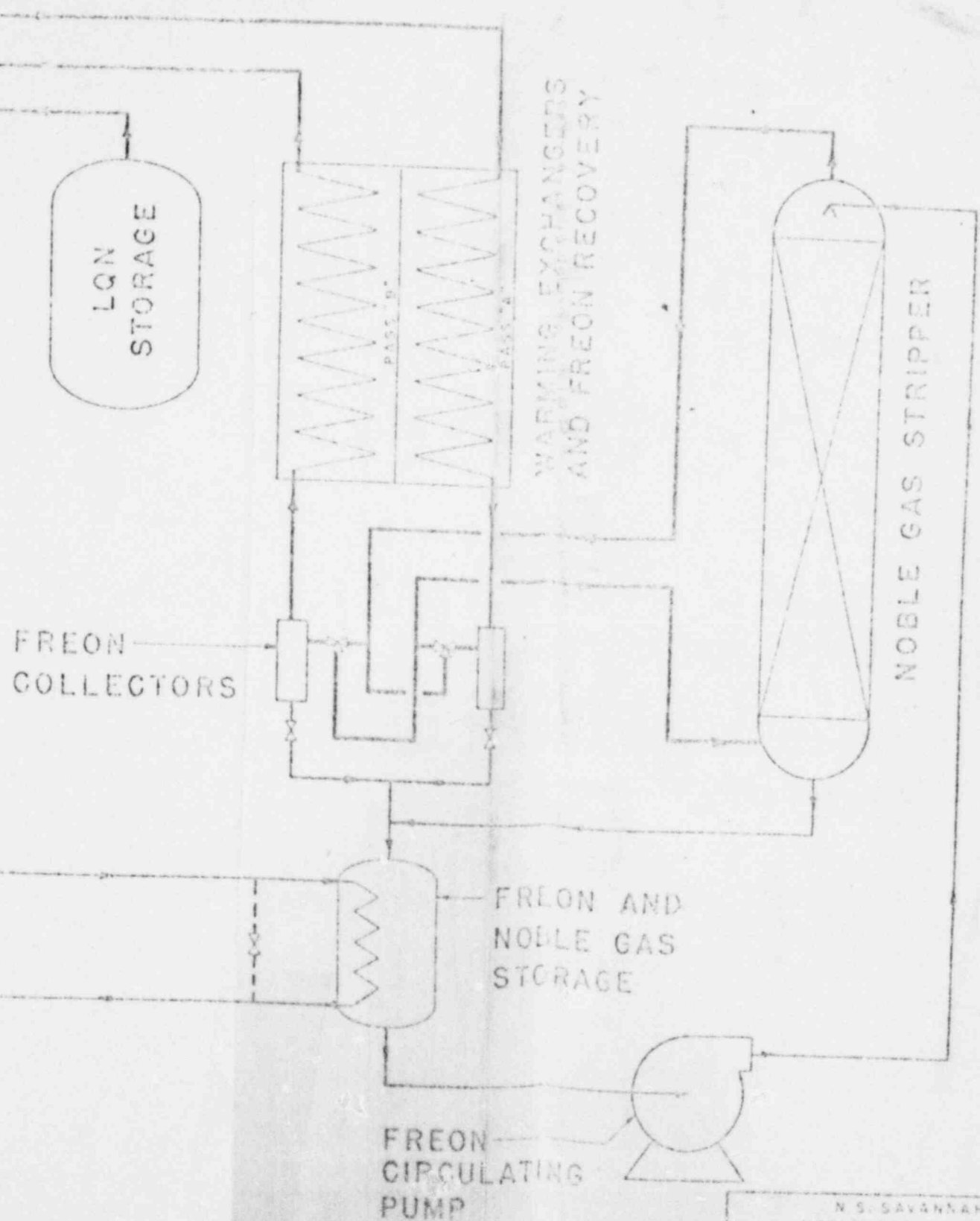
U.S. GOVERNMENT  
 WORKING MODEL  
 SYSTEM C  
 CONTAINMENT SYSTEM  
 CONTAINMENT SYSTEM  
 CONTAINMENT SYSTEM



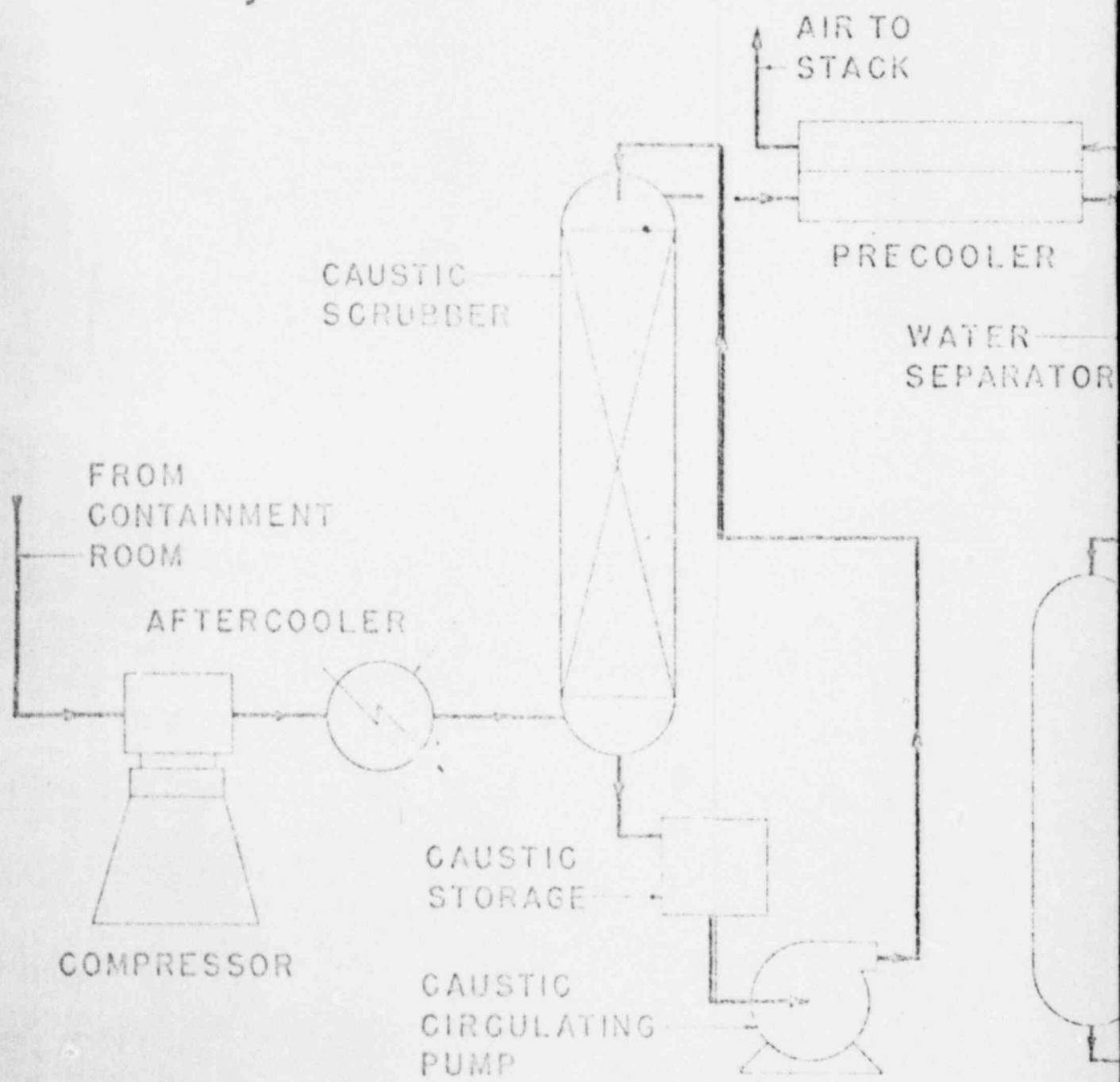
N. S. SAVANNAH  
 NOBLE GAS RECOVERY  
 SYSTEM D  
 STATIC FREON ABSORPTION  
 FLOW DIAGRAM  
AIR REDUCTION SALES COMPANY





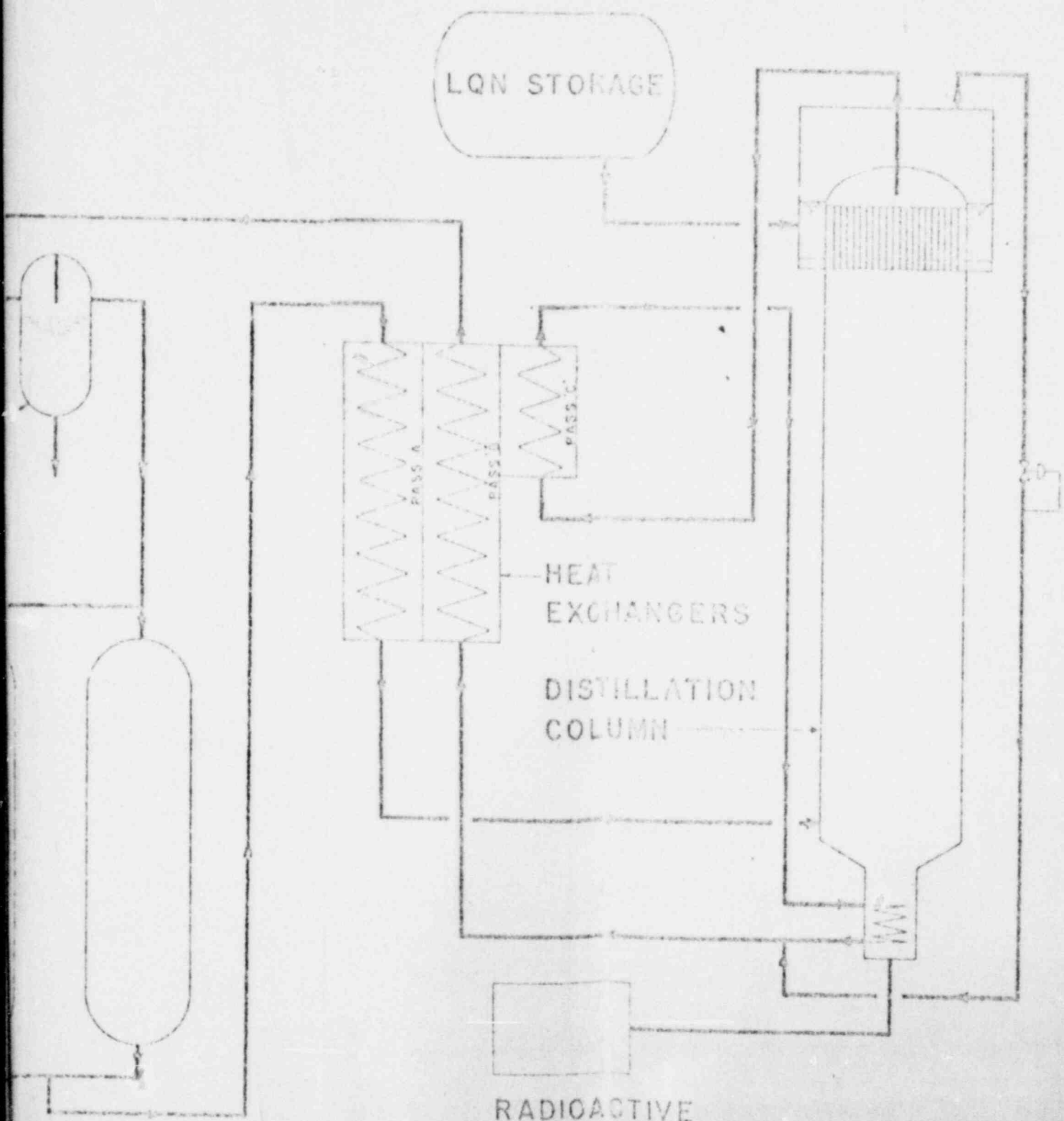
N. S. SAVANNAH  
 NOBLE GAS RECOVERY  
 SYSTEM E  
 DYNAMIC FREON ABSORPTION  
 FLOW DIAGRAM  
 AIR REDUCED BY JALAS COMPANY





S

DATE	BY	NO.



CA GEL  
TOWERS

LQN STORAGE

PASS A  
PASS B  
PASS C  
HEAT EXCHANGERS

DISTILLATION COLUMN

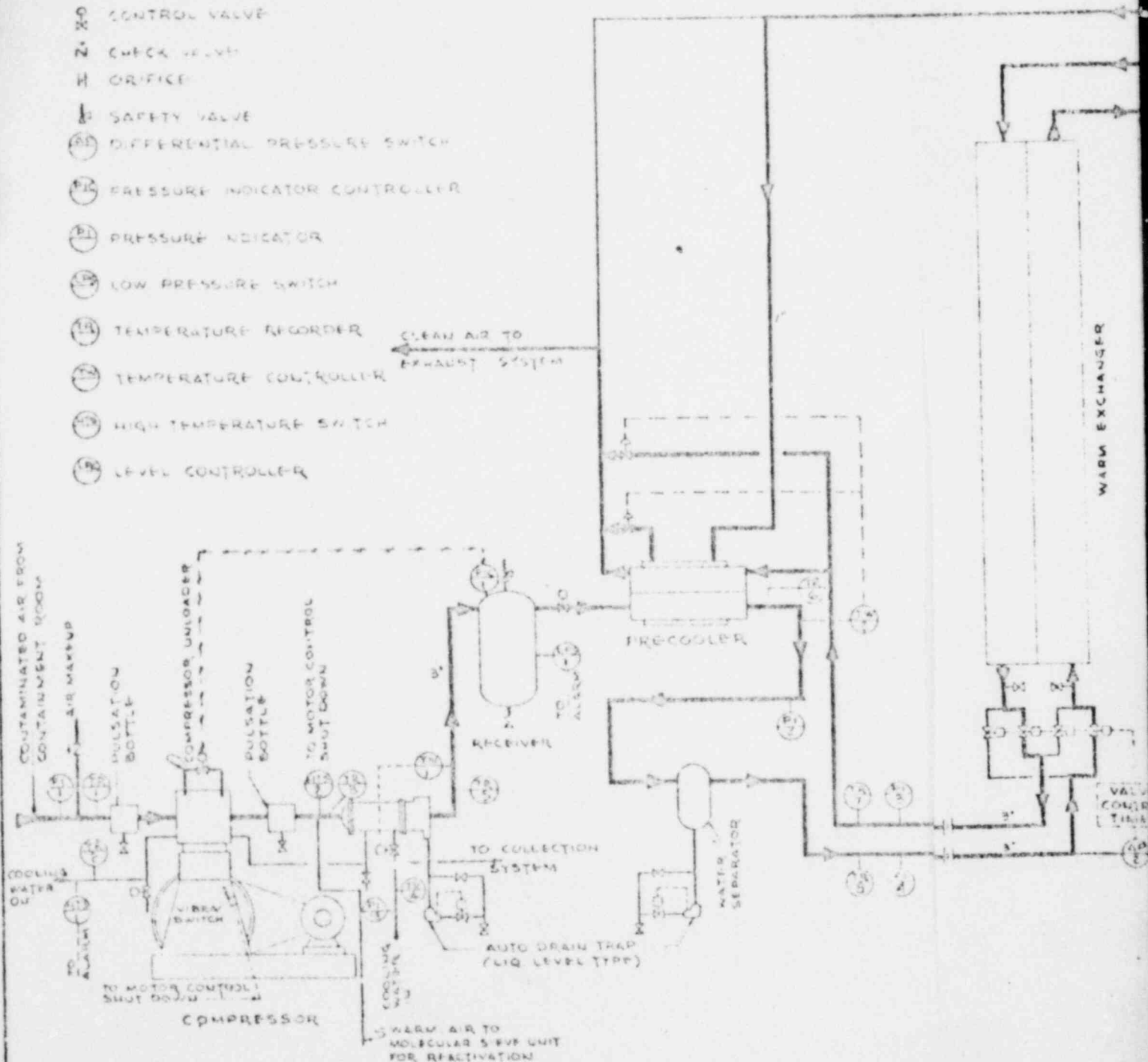
RADIOACTIVE MATERIALS STORAGE SYSTEM

N. S. SAVANNAH  
NOBLE GAS RECOVERY  
SYSTEM F  
NON REVERSING PROCESS  
FLOW DIAGRAM

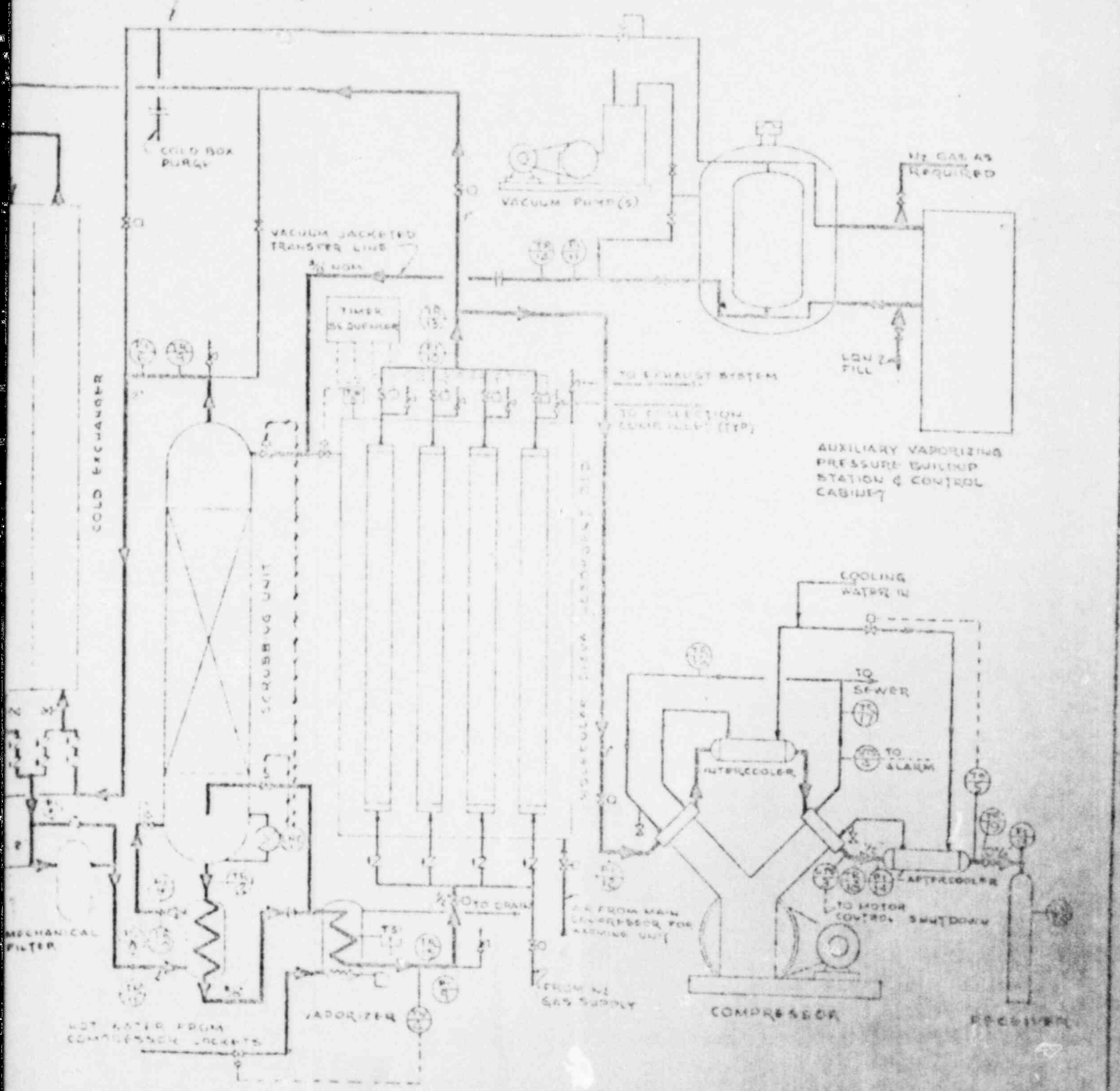
REVISION	DATE	BY	REASON

### LEGEND

- Z MANUAL SERVICING VALVE
- RC REMOTELY ACTUATED VALVE
- XO CONTROL VALVE
- Z CHECK VALVE
- H ORIFICE
- SV SAFETY VALVE
- PS DIFFERENTIAL PRESSURE SWITCH
- PI PRESSURE INDICATOR CONTROLLER
- PI PRESSURE INDICATOR
- LS LOW PRESSURE SWITCH
- TR TEMPERATURE RECORDER
- TC TEMPERATURE CONTROLLER
- HS HIGH TEMPERATURE SWITCH
- LC LEVEL CONTROLLER



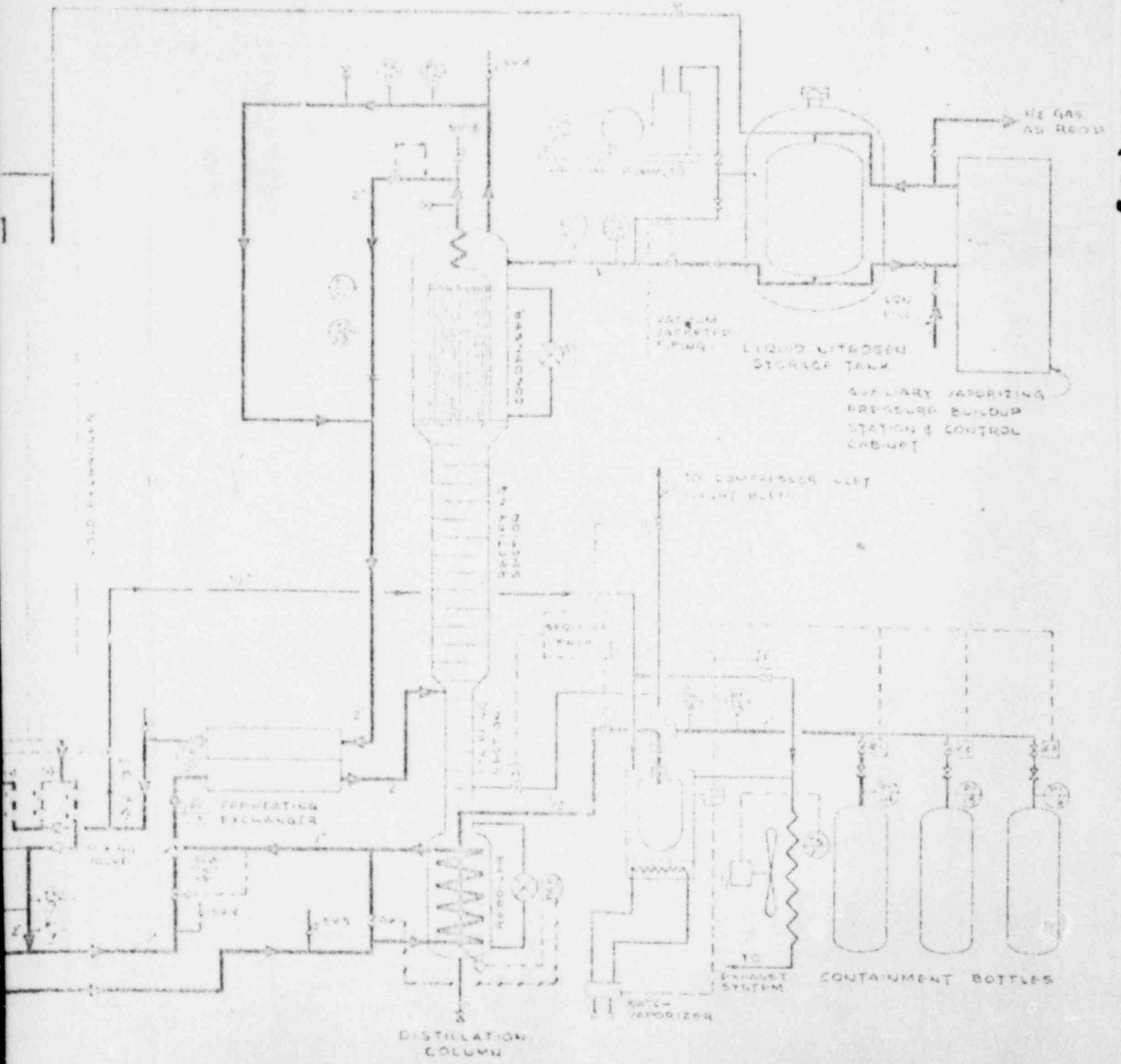
DATE	BY	NO.



AS SAVANNAH  
 NOBLE GAS RECOVERY  
**SYSTEM B**  
 SOLID ADSORPTION WITH  
 CRYOGENIC PURIFICATION  
 FIG. 10-2 INSTRUMENT DIAGRAM  
 AIR PRODUCTS GASES COMPANY  
 A DIVISION OF AIR LIQUIDE COMPANY INC.  
 ALL RIGHTS RESERVED





HS SAVANNAH  
 NOBLE GAS RECOVERY  
**SYSTEM C**  
 CRYOGENIC DISTILLATION  
 PIPING & INSTRUMENT DIAGRAM  
 AIR REGISTRATION SALES COMPANY

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*Welding, Cutting, Steel Conditioning and Cryogenic Equipment*

*Cryogenic Equipment:*

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#### HOFMAN LABORATORIES

225 Parkhurst Street, Newark, N. J. 07114

#### AIR REDUCTION PACIFIC COMPANY

100 California Street, San Francisco, Calif. 94111

*Industrial Gases and Cryogenic Liquids*

*Welding, Cutting, Steel Conditioning and Cryogenic Equipment*

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*Calcium Hydroxide*

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#### AIRCO PLASTICS PRODUCTS

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*Hungarford Molded Vinyl Plastics Products*

*Ductall Flexible Plastic Hose*

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1400 East Washington Avenue, Madison, Wis.

*Medical Gases and Volatile Anesthetic Agents*

*Hospital Equipment*

*Pharmaceuticals*

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*Ferrous Alloys and Metals*

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*Carbon Dioxide and Related Equipment*

#### SPLIT CARBON COMPANY

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*Electronic Components*

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*Carbon Dioxide*

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*Hospital Equipment*

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