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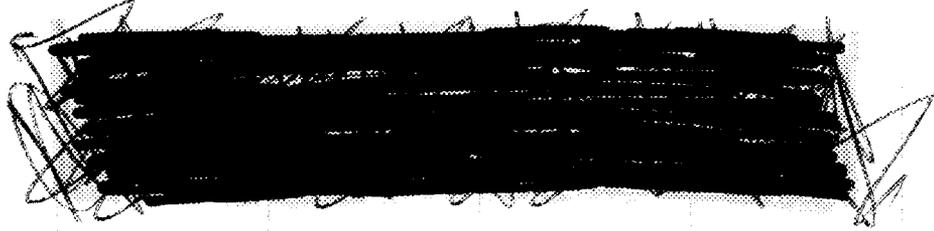
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# EFFLUENT CONTROL IN FUEL REPROCESSING PLANTS

O. O. Yorbre  
F. E. Harrington  
D. S. Joy



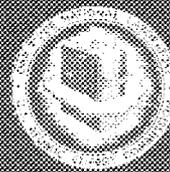
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EFFLUENT CONTROL IN FUEL REPROCESSING PLANTS

O. O. Yarbro, F. E. Harrington, and D. S. Joy

MARCH 1974

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## CONTENTS

	<u>Page</u>
Abstract . . . . .	1
1. Introduction . . . . .	1
2. Summary and Recommendations. . . . .	3
2.1 Reduction in Volume of Effluents. . . . .	3
2.2 Avoidance of Bypassing of Contaminant Trapping Systems. . . . .	4
2.3 Tritium Containment . . . . .	5
2.4 Krypton Containment . . . . .	7
2.5 Iodine Containment. . . . .	7
2.6 Particulates. . . . .	9
2.7 Recommendations . . . . .	10
3. Containment Philosophy . . . . .	11
3.1 Minimization of Routine Effluent Volumes. . . . .	11
3.2 Containment During Maintenance. . . . .	13
3.3 Decommissioning. . . . .	14
4. Process Description. . . . .	17
4.1 Flowsheet for "Near Zero" Release . . . . .	17
4.2 Tritium Control . . . . .	19
4.3 Krypton-Xenon Control . . . . .	21
4.4 Iodine Control. . . . .	23
4.5 Recovery and Recycle of Acid and Water. . . . .	25
5. Modeling and Computer Analysis . . . . .	26
5.1 Distribution and Removal of Tritium . . . . .	29
5.2 Distribution and Removal of Krypton . . . . .	32
5.3 Distribution and Retention of Iodine. . . . .	35
5.4 Distribution of Other Fission Products. . . . .	47
6. References . . . . .	47
7. Appendix . . . . .	49



EFFLUENT CONTROL IN FUEL REPROCESSING PLANTS

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## ABSTRACT

This report summarizes a study of how extrapolations of current and developing technology might be applied to the task of reducing radioactive effluents from future fuel reprocessing plants to "near zero." The study has indicated that very significant reductions of effluents can be achieved by integrating advanced effluent control systems with new concepts of containment and ventilation that would reduce net inleakage of air to the process enclosures and provide for extensive recycle of gases and liquids.

If net plant effluent flow rates can be thus reduced and if the highly efficient fission product removal systems, under development, do in fact become available, it should be possible to reduce the discharge of activity to the environment in future plants by four to six orders of magnitude. Overall plant retention factors in the order of  $10^{10}$  for iodine,  $10^5$  for tritium and krypton, and  $10^{14}$  to  $10^{16}$  for particulates appear possible.

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1. INTRODUCTION

An increasing interest in protecting the environment is necessitating the reevaluation of systems for effluent control at future fuel reprocessing facilities, particularly those for processing LMFBR fuels. In the past, radioactive discharges have been limited to quantities that would yield concentrations of radioactive contaminants at site boundaries well below levels set by national and international agencies for continuous intake by the public. The present emphasis is to ensure that releases of radioactivity are, in addition, kept to as low a value as practicable. Thus, effluent control systems that are capable of minimizing the impact of all effluents on the environment are of interest.

As IMFBRs, with higher burnup levels, higher specific power, and an economic incentive to reduce preprocessing decay time, assume their projected role in the power economy of the future, the input level of fission products to reprocessing plants will increase significantly. This higher input level of activity, coupled with possible reductions in the permissible release of activity to the environment, will place very stringent demands on the effluent control systems and require advanced processes for the control and removal of the volatile fission products from effluent streams.

The objective of this study was to incorporate advanced systems into a containment and operating philosophy directed at reducing the total effluent volume, and activity, from a reactor fuel reprocessing plant to as low a value as possible. Although some of these systems may have application in reducing effluent activity from present generation reprocessing plants, their full potential can only be realized when they are applied to new plants employing radically different containment and ventilation philosophies. This study deals specifically with the advanced plant designed to minimize overall effluent volumes and activity. Other studies<sup>1</sup> have addressed the problem of applying some of these same advanced effluent control systems to present-generation plants.

The primary objectives of the "Fuel Reprocessing Plant Effluent Control Study" can be summarized as follows:

1. To define, in semiquantitative terms, the significant sources and the distribution of fission product activity throughout the reprocessing plant system for a range of effluent control systems.
2. To determine the optimum location and required decontamination factors for fission product removal systems on both gaseous and liquid process streams as they relate to required overall plant retention factors.
3. To indicate areas where insufficient data are available for accurately predicting the behavior of important fission products in a process or effluent control system, and to indicate

requirements for further research and development to eliminate such data gaps.

As a basis for the study, it was assumed that highly efficient removal systems for all of the significant fission products do become available and that improved containment at greatly reduced off-gas rates is attainable. Very little actual equipment conceptual design was done in connection with this study, and no detailed cost estimates were made. We believe, however, that reductions in effluent activities and volumes to levels approaching "near zero" could be achieved in future plants.

The "near-zero" confinement objective can be realized by a reasonable projection of the technology presently in development. The added cost associated with "near zero" confinement may increase current processing costs between 50 and 100%.

## 2. SUMMARY AND RECOMMENDATIONS

Because the related process development work is not complete, it is not possible to accurately determine the practical limit for fission product retention. However, based on the assumptions used in this study it appears that the following retention factors can be attained: iodine -  $10^{10}$ , noble gases and tritium -  $10^5$ . Variations from these values will depend on the success of the development programs now underway, and on the amount of effort that is deemed practical to expend in retaining radioactive substances.

Key factors in reducing the quantity of radioactivity released to the environment are discussed below:

### 2.1 Reduction in Volume of Effluents

The practical extent of treatment of an effluent is determined in large measure by the volume of the effluent to be treated. Also, reduction of the content of a substance below a certain minimum concentration is apparently very difficult through use of the means now available or under development. The recycle of the cell atmosphere, non-reactant gas

used for process functions, water, acids, and other process fluids is essential if a substantial reduction is to be realized in the total release of radioactivity from a reprocessing facility. This degree of recycle has never been attempted in an aqueous reprocessing system. Because it has never been attempted on all streams and over a protracted period, we can only speculate about the problems to be encountered. The potential problem of greatest concern in recycle of liquids (primarily water and nitric acid) is a reduction in decontamination or separation effectiveness owing to a decrease in purity of the fluid being used to extract (or scrub) a given material from another fluid. A variation of that potential problem is the introduction of an interfering substance via a recycle fluid. Development work to date indicates that offending materials can be removed from recycle streams to exceedingly low levels.

The principal problem concerning recycle of process gases and cell atmosphere is related to the lower practical limit on inleakage to the cells, and from the cells into the process equipment. Aqueous reprocessing facilities have traditionally operated with large net flows of air into cells and certain process vessels. A large shielded fuel examination facility (the High-Level Fuel Examination Facility at the National Reactor Testing Station, Arco, Idaho) is operating with an air inleakage rate of 0.004 cfm; this facility was constructed at a cost of about \$10 million. We believe that a practical inleakage rate for a reprocessing facility built with the intent of obtaining a low air inleakage rate is 100 cfm or less. We believe that it is practical to design, construct, and operate process equipment, including the shear and dissolver, such that the total inleakage to the head-end process equipment will not exceed 100 cfm.

## 2.2 Avoidance of Bypassing of Contaminant Trapping Systems

A trapping system obviously cannot decrease the activity of a stream it does not treat; however, practical considerations may result

in a stream having either very low contaminant content, or else very low probability of contamination, being routed to the environment without full treatment. Careful planning of the effluent treatment train is required to avoid, or reduce the effects of, this limitation on trapping effectiveness.

The processes that appear to have the greatest potential for practical application in reducing radioactive emissions from a reprocessing facility are yet being developed. The processes under development for iodine, noble gas, and tritium retention are described below.

### 2.3 Tritium Containment

The containment of tritium in a fuel reprocessing plant can be achieved by head-end evolution and trapping (voloxidation), total water containment, or a combination of both methods. The overall plant containment factor for tritium that can be achieved by head-end evolution and trapping is limited by the inability to totally evolve tritium from the fuel as well as by the escape of small quantities of tritium from failed fuel during storage and handling. A practical upper limit on tritium containment factors, based on head-end evolution and trapping, is of the order of 100. If water containment is used to control tritium release, an overall plant containment factor in excess of  $10^4$  appears to be achievable if the total plant off-gas rates are of the order of 100 cfm and the total water input to the plant is kept sufficiently low for long-term accumulation or permanent disposal. Specific case studies reported in Sect. 5.1 indicate overall tritium containment factors in the range of  $10^8$  for combined voloxidation and water retention, assuming a final off-gas dewpoint of  $-140^\circ\text{F}$  and tritium removal by a factor of 200 across a final krypton removal system. With a more practical dewpoint of  $-100^\circ\text{F}$  and no credit for the final krypton system, overall tritium retention in the range of  $10^5$  to  $10^6$  appears possible. Typical plant retention factors for the three systems of tritium control as a function of total plant effluent gas are summarized in Table 2.1.

Table 2.1. Summary of Tritium Retention Factors

System for Tritium Control	Plant Off-Gas Rate (scfm)	Probable Attainable Tritium Retention Factor
Head-end tritium evolution and trapping	100	100
	1000	100
Plant water retention and recycle (-100°F dewpoint on plant off-gas)	100	$10^4$
	1000	$10^3$
Combined trapping and water retention	100	$10^6$
	1000	$10^5$

## 2.4 Krypton Containment

Krypton is totally evolved during the dissolution step; however, some escape and evolution occur during preceding steps. As in the case of tritium, any escape and bypass of krypton around the primary removal system will set an upper limit of the effectiveness of primary control. Locating the primary krypton removal system on the combined dissolver and head-end off-gas, including the total off-gas from the storage and mechanical head-end cell, should minimize the bypass of krypton, and overall retention factors for krypton in the range of  $10^3$  should be attainable based on primary evolution and trapping. If retention factors in excess of  $10^3$  are desired, the bypass of trace quantities of krypton around the primary system becomes a serious limitation. If the cell off-gas flow is limited to a few hundred cubic feet per minute, it is practical to install krypton removal equipment on the total cell off-gas stream, either instead of or in addition to primary treatment. If primary removal is omitted and krypton is removed only from total plant effluent gas, overall retention factors in the range of  $10^3$  to  $10^4$  should be attainable; however, the cell atmosphere would contain a large krypton inventory from recycle process gas. If overall krypton retention factors much in excess of  $10^3$  are desired, a combination of primary krypton retention combined with krypton removal from total plant off-gas should be capable of yielding overall retention factors in the range of  $10^5$  to  $10^6$ . Detailed results for a number of case studies for krypton are summarized in Sect. 5.2.

## 2.5 Iodine Containment

The behavior of iodine in the process and off-gas systems is very complex, with interaction occurring between the numerous systems associated with iodine control. The process systems directly associated with iodine control include the following:

- (1) Iodine evolution (dissolver solution).
- (2) Primary iodine removal (dissolver off-gas).
- (3) Secondary iodine removal (vessel off-gas).

- (4) Iodine removal from recycle water and acid.
- (5) Final off-gas treatment (plant effluent).

Other systems having a significant effect on iodine behavior include the high-level waste handling system, which may revolatilize iodine into the off-gas system, and the final krypton removal systems, which effectively removes iodine from the off-gas.

With all iodine control systems operating at typical efficiencies, and a total plant off-gas in the range of 100 cfm, overall plant iodine retention factors in excess of  $10^{10}$  appear to be attainable. This assumes that the various iodine removal systems can be operated in series without loss of efficiency. Experiments have shown this to be true for DFs of the order of  $10^6$ ; however, it will require pilot operation with short-decayed fuel to demonstrate overall DFs of  $> 10^{10}$ . The results of individual case studies for iodine are presented in Sect. 5.3.

The relative effectiveness of the various iodine removal systems is detailed in Sect. 5 and tabulated in Table 5.8. In general, the effective DF of the primary iodine removal system is limited to  $\sim 100$  by iodine bypass around the system; thus there is little incentive in providing an actual primary system DF much in excess of  $10^3$  for iodine control purposes. In the case studies summarized in Table 5.8, primary system DFs in the order of  $10^7$  are listed. This was done to minimize the mass of iodine in the feed to krypton removal, and probably is not required.

Secondary iodine removal was assumed to yield a DF in the range of  $10^4$ , which was effective in reducing downstream iodine concentrations. It should be possible to increase this DF at least an order of magnitude if desired.

The effective iodine DF across the vessel off-gas and final off-gas treatment systems was limited in most of the case studies listed in Sect. 5.3 by the iodine content of recycle acid and water used in the final off-gas treatment steps. In general, the overall plant iodine retention factor was limited to  $\sim 10^{11}$ , regardless of the individual efficiencies of the final iodine removal systems. Additional improvement in iodine retention must come from further purification of recycle water.

The effect of omitting various sections of the overall iodine control system was studied in order to obtain a rough estimate of the relative value of individual systems as well as an indication of the effect of short-term failure of one of the systems. Eliminating the primary iodine removal system caused a decrease of one to two orders of magnitude (runs 24 through 27) in overall plant retention of iodine. Other undesirable effects of not operating the iodine evolution and primary removal systems include a much higher inventory of iodine throughout all process steps and, in particular, in the waste handling systems. It is not recommended that the iodine evolution and primary iodine removal systems be omitted if overall plant retention factors in excess of  $10^3$  are desired.

If the secondary iodine removal system is not operated, a loss in DF by factors ranging from  $4 \times 10^3$  to  $3 \times 10^4$  (runs 28 through 31) is indicated. In addition to a large loss of overall iodine retention, the iodine content of gas recycled for cell and process use increased by three to four orders of magnitude.

If the iodine removal efficiency of the recycle water and acid purification systems is reduced from 99% to 90%, a loss of overall iodine retention by one to two orders of magnitude results. The primary effect here is the recontamination of off-gas streams by iodine-containing water used in scrubbers and other gas treatment equipment.

The iodine removal system associated with the final off-gas treatment does not have any effect on previous steps, and the loss of overall DF is equal to the actual DF of the final system, which was assumed to be  $10^3$  in this study. Additional decontamination was obtained in cases where a secondary krypton system was assumed to treat the final off-gas stream prior to its discharge to the stack.

## 2.6 Particulates

Removal of particulate activity from effluents to levels four to five orders of magnitude below current practice should be possible by an extrapolation of current technology. The off-gas from the vessel

off-gas treatment system for the "near zero" release concept should be similar in particulate content to the off-gas from existing plants. Due to the much lower volume, extensive treatment for particulate removal in the final off-gas treatment system should yield the desired results. This, however, is an area where further research and development is required to demonstrate the efficiencies of ultrahigh efficiency filters at low flow rates.

## 2.7 Recommendations

We recommend that the development program to reduce radioactive emissions to the environment be continued and that it include the following items:

- (1) Continuance of existing laboratory programs aimed at separating fission products from effluent and process streams.
- (2) Preparation of a conceptual design of an advanced model plant for reprocessing of LMFBR fuels on a pilot plant scale.
- (3) Design and cold development of process equipment components and unit process equipment assemblies.
- (4) Integration of the various process systems into an operating cold pilot plant. This facility should be complete with containment barriers and ancillary equipment to resolve problems of interfacing, recycle streams, and process control.
- (5) Operation of the above systems in a shielded facility for an extended period of time using irradiated fuel as feed material to the plant. In time, the radioactivity of the feed material should reach or exceed the peak level expected in a commercial facility.
- (6) Communication with commercial reprocessors, especially in the early stages of development, to assure that their experience is used in determining the direction of the

program, and to ensure that the information which is obtained is adequate for plant design and operation.

### 3. CONTAINMENT PHILOSOPHY

In order to meet the objectives of minimizing all activity releases from a fuel reprocessing plant, a basic understanding of the interactions of the process steps is required. The functional process steps must be integrated into an off-gas and containment philosophy which minimizes effluent volumes and makes efficient and economical application of advanced off-gas and liquid effluent treatment systems possible. A high degree of overall containment must be maintained during all phases of plant life, including routine operation, maintenance, and the decommissioning phase at the end of a plant's useful life. Each of these operations has its own unique problems, which are outlined in general terms in this section; however, each will require an in-depth study, including conceptual design and some engineering demonstrations to ensure their compatibility with the overall objective of "near zero" release.

#### 3.1 Minimization of Routine Effluent Volumes

The key to reducing the activity in effluents from a fuel reprocessing plant to very low levels lies in decreasing the volumes of both gaseous and liquid effluents to as low a volume as practical. The current practice of using once-through ventilation at rates in the 100,000 cfm range for cell enclosures is not compatible with the "near zero" release of activity from the plant. Removal of trace concentrations of tritium, krypton, and iodine from very large gas flows is economically infeasible as well as technically unsound at the present time.

The reduction of effluent volumes by orders of magnitude will require new approaches to cell containment and ventilation. One concept directed toward minimal effluent volumes is illustrated in Fig. 3.1. The equipment is housed in sealed-cell enclosures, which limits net inleakage of air to minimal volumes. In the ideal case this inleakage rate would be sufficiently small (<100 cfm range) to be accepted directly into the equipment

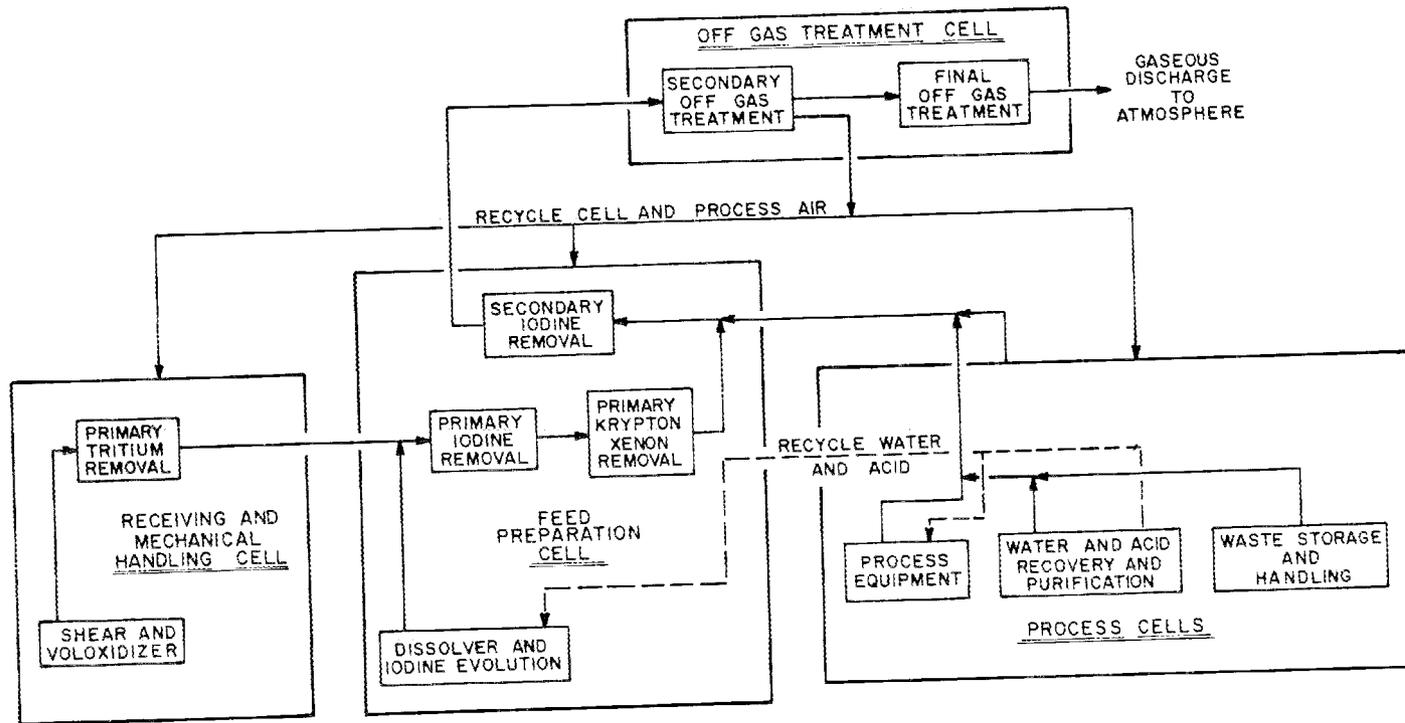


Fig. 3.1. Fuel Reprocessing Plant-Effluent Control Flowsheet.

off-gas system. In order to further minimize effluent volumes, process liquid and air requirements are supplied by recycle systems. Only the small volume of gas representing the net inleakage to the cell enclosure is given final treatment and released to the atmosphere. The small net liquid input to the plant is decontaminated from all activity other than tritium, and then permanently stored or safely disposed of to prevent contamination of the environment.

### 3.2 Containment During Maintenance

Maintenance is an integral part of long-term plant operation and must be carried out in a manner consistent with overall containment requirements and objectives. The approach taken in current installations is to decontaminate equipment and cells, as required, to a level compatible with the maintenance approach to be used. Cell ventilation rates are maintained at or above normal flows and passed through filters to effect particulate removal. Such an approach is acceptable for current-generation plants operating on long-cooled fuel, and with currently accepted discharge standards. The required retention factors for particulates can be maintained by normal filtration equipment. Present standards do not stipulate retention of krypton or tritium, and the required overall iodine retention factor of  $10^3$  to  $10^5$  can easily be met by a combination of the long fuel decay time and a modest amount of equipment flushing and decontamination prior to maintenance.

The situation changes significantly for the "near zero" release concept of plant operation from at least two aspects. The normal off-gas flow rate from the cell enclosure is orders of magnitude below present practice and far below the rate required for once-through ventilation for "open" cell maintenance. Off-gas treatment equipment designed for the highly efficient removal of contaminants such as particulates, iodine, tritium, and krypton cannot accommodate the high flow rates required for once-through ventilation. Decontamination of equipment and of cells to the levels required for high overall retention factors for iodine ( $>10^6$ ) and tritium ( $>10^3$ ) does not appear practical. In this application, a containment concept which maintains the sealed-cell philosophy with low

off-gas rates during maintenance appears appropriate. For intermediate retention factors a containment concept based on low cell off-gas with extensive treatment during normal operations and a once-through ventilation for maintenance, coupled with intensive decontamination and iodine removal, may be practical.

One approach to a sealed-cell maintenance concept is illustrated in Fig. 3.2. Under normal operating conditions the cell is sealed and shielded from the crane bay and the maintenance area, and the total air inleakage to both areas is removed as cell off-gas. During maintenance operations the shield blocks over the cell are removed, as required, and a recirculating air cleaning system provides a relatively clean air supply to the crane bay area and furnishes an air sweep into the cell area. It should be possible for personnel wearing air suits to enter the crane bay with the cell sealed off if this becomes necessary in order to accomplish crane maintenance or similar operations.

### 3.3 Decommissioning

A detailed analysis of all the aspects of the decommissioning of a fuel reprocessing plant is beyond the scope of this report and represents a needed study in its own right. Presented here is an outline of some of the options related to the decommissioning of a plant, designed and operated during its useful life to minimize the overall effects of effluents on the environment. A number of possible objectives can be considered as the termination point of a decommissioning program and include the following:<sup>2</sup>

- (1) Thorough decontamination of the facility, with subsequent reuse for other nuclear (or possibly nonnuclear) functions.
- (2) Thorough decontamination, after which the cell and building are sealed to prevent access by the public.
- (3) Thorough decontamination, followed by removal of equipment and structures, with restoration of the land to some form of controlled usage.

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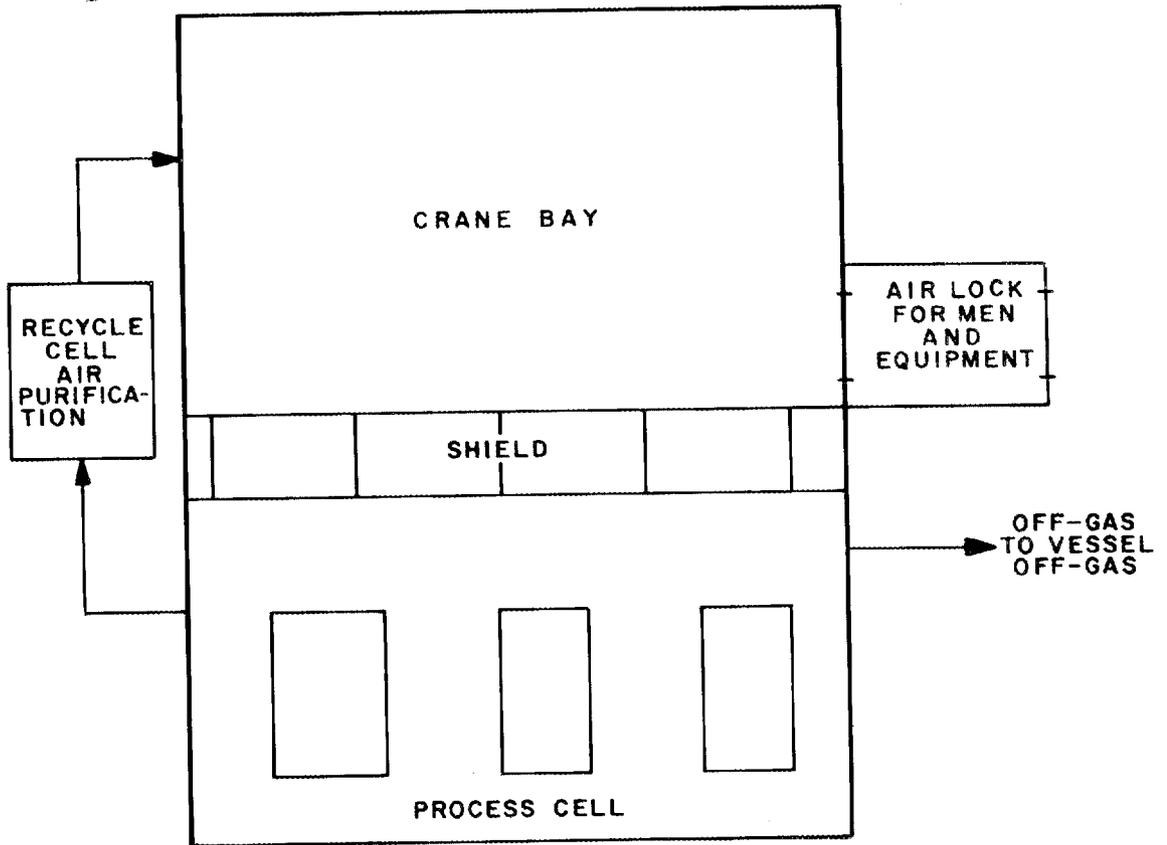


Fig. 3.2. Sealed Cell Maintenance Concept.

To be consistent with the "near zero" release philosophy, the discharge of activity to the environment during and following the decommissioning phase should not exceed those levels experienced during the useful life of the plant.

The process of decommissioning progresses through a series of events, including the following major areas of endeavor:

- (1) Decontamination of equipment, piping, and cell interior.
- (2) Conversion of all liquid wastes to solids for disposal, with recovery and purification of water and acid inventories.
- (3) Disposal of water and acid inventories.
- (4) Disposition of building structure by one of the methods outlined above.

The first two tasks, which include decontamination of equipment and solidification and disposal of waste, would be accomplished through procedures normally used for such operations during the useful life of the plant. Disposal of the water and acid inventories poses unique problems, particularly if large amounts of tritium have been retained during the life of the plant in the water and acid. The plant inventory of tritium might range from 10 to 1000 days' total input, depending on the type and degree of tritium control applied during operation of the plant. For the case where a tritium retention factor of  $10^3$  or greater was realized (either wholly or partially by minimizing water release), the inventory would be of the order of a few hundred days' input, making controlled discharge an impractical method of water disposal.

Possible methods for disposing of the acid inventory include the following:

- (1) Transfer for use to another processing facility in the early stages of hot startup.
- (2) Long-term liquid storage for tritium decay.
- (3) Conversion to a solid form for permanent disposal with recovery of the water.

Similarly, the water inventory in the plant may be disposed by one of the first two methods listed above, as well as by the following possible alternatives:

- (4) Conversion to concrete or similar hydrated solid for permanent disposal.
- (5) Disposal to some geological formation or area with retention time measured in hundreds of years.
- (6) Isotopic separation and concentration of the tritium, with subsequent release of the decontaminated water.

Once the decommissioning program is complete, the long-term effect of the site on the local environment should be minimal.

#### 4. PROCESS DESCRIPTION

##### 4.1 Flowsheet for "Near Zero" Release

In order to meet the objectives of the "near zero" release concept, a basic understanding of the interactions between the various process steps that take place in a reprocessing plant is required. Of particular importance is a knowledge of the behavior of all significant fission products throughout the process system. For modeling purposes, a plant flowsheet (Fig. A-1 in the Appendix) depicting 24 functional process steps located in eight containment cells was developed, and the interactions between process and effluent treatment functions were defined. A simplified flowsheet retaining sufficient detail to illustrate important effluent control features is shown in Fig. 3.1.

Fuel elements are introduced into a receiving and storage cell through a seal arrangement which prevents inleakage of air into the cell. A small fraction of the volatile fission products will escape to the cell environment from damaged fuel elements during storage and open handling of the elements in the cell. Ideally, all off-gas from the receiving and mechanical handling cells would be routed through the primary tritium, iodine,

and krypton removal systems. Such an approach is practical only if the total gas inleakage to the cell is sufficiently small to be accommodated in the primary off-gas treatment systems. A major portion of the tritium and significant quantities of iodine and krypton are evolved in the shear and voloxidation steps, and the resulting off-gas is routed through the primary tritium removal equipment.

The remainder of the krypton and most of the iodine are evolved during dissolution and feed adjustment, and the off-gas from these functions is combined with the effluent from the tritium removal system and passed successively through the primary iodine removal and the primary krypton removal systems. A fraction of a percent of the total tritium and as much as 1% of the iodine may follow the feed into solvent extraction and subsequently be released to the off-gas system in the process steps downstream. This material, coupled with any additional bypass around the primary systems from the head-end cells, limits the maximum containment factor that can be achieved by primary treatment.

Fission product behavior in the main-line process equipment is important because these systems represent sources of volatile or particulate activity into the off-gas system. Off-gas from all equipment downstream of the feed adjustment step is combined with the off-gas from primary treatment and passes through secondary treatment. The secondary off-gas treatment system contains filters for particulate removal and scrubbers for nitrous oxides, iodine, and ruthenium removal. A major part of the gaseous effluent from secondary treatment is recycled for process use; a relatively small fraction is treated further and then discharged to the atmosphere. The final off-gas treatment includes provisions for the removal of particulates, iodine, and, if required, tritium and krypton.

One of the main features of the "near zero" release concept is the use of recycle to reduce effluent volumes of both gases and liquids. The recovery and purification of acid and water for recycle are indicated. Ideally, there would be no net discharge of liquids, and any excess water would be converted to a solid form or permanently stored. If a water discharge is required, the relatively small volume would be extensively purified and subsequently released as a vapor to a stack.

This section is intended to give an overall flowsheet description; more detailed information on the important aspects of effluent control are covered in subsequent sections.

#### 4.2 Tritium Control

The control of tritium in a fuel reprocessing plant based on head-end evolution and trapping is illustrated schematically in Fig. 4.1. Some tritium is released as the fuel elements are sheared, and most of the remainder is evolved during the voloxidation step, in which the sheared fuel is heated to 450°C in air for 2 to 4 hr. The combined off-gas from the shear and voloxidizer is passed over an oxidation catalyst to convert all the tritium to water; then the tritiated water is trapped from the gas stream with a desiccant such as molecular sieves. This type of approach to tritium control should be capable of providing overall plant retention factors for tritium in the order of 100, limited by the residual tritium in the fuel after voloxidation and any bypass of tritium around the tritium removal system. The volume of water that must be stored as tritium waste is dependent on the extent to which water can be excluded from the voloxidation and shearing equipment. The minimum volume of accumulated water is set by the volume of off-gas that is treated and the water removal capabilities of the desiccant used. The major unresolved questions relative to this approach to tritium control include the mechanical aspects of operating a voloxidizer while maintaining temperature control and good containment with low off-gas volume. In contrast to other equipment for reducing radioactive emissions, voloxidation equipment may occupy a large amount of cell space and be unusually expensive.

A process flowsheet for a typical tritium removal system using molecular sieves is summarized in the Appendix (Fig. A-2).

An alternative approach to tritium control in a "near zero" release plant containment concept consists of the nearly total retention ( $< 3 \times 10^{-7}$  of total water inventory released per day for an overall tritium retention factor of  $10^3$ ) of water within the plant. In order for this to be a practical approach, the total plant off-gas flow rate must be

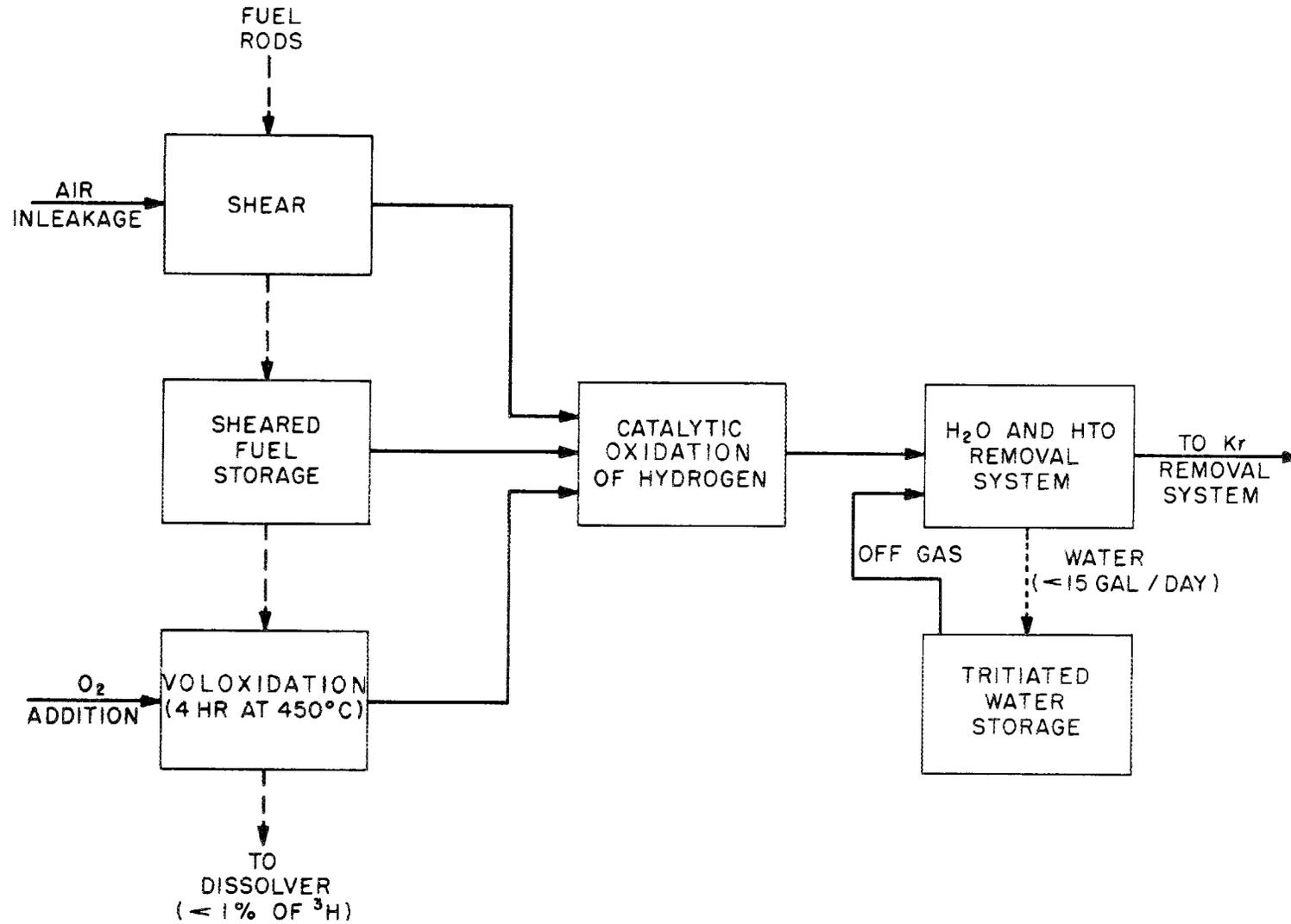


Fig. 4.1. Flowsheet for Tritium Removal.

in the range of 1000 cfm or less and all net water additions over the life of the plant must be retained. Although such a tritium retention system is technically simple, tritium in the plant water inventory will cause minor problems in personnel protection and at the termination of plant operation will pose a disposal problem (see Sect. 3.3).

#### 4.3 Krypton-Xenon Control

The retention of krypton will be limited by the ability to route all krypton-containing streams to the primary krypton removal system. The major sources of krypton include the dissolver off-gas and the exit gas streams from the tritium removal system. As a trace of krypton will be present in the off-gas from the storage and handling cell, the off-gas from this cell should be included in the feed to the primary krypton removal system. The combined krypton-containing off-gases are passed successively through the primary iodine removal and the primary krypton-xenon removal systems (Fig. 4.2). Overall plant retention factors for krypton of the order of  $10^3$  should be obtainable, based on primary krypton retention, if all head-end cell and equipment off-gases are routed to primary treatment.

Krypton<sup>3</sup> can be effectively removed from an off-gas stream by absorption in liquid nitrogen or by scrubbing with a liquid fluorocarbon such as refrigerant-12. The liquid nitrogen scrubbing system has been successfully demonstrated for a number of years at the Idaho Chemical Processing Plant, and similar systems are marketed commercially for the removal of krypton-xenon from reactor off-gas. The feed gas for the cryogenic system must be free of all contaminants such as CO<sub>2</sub>, NO<sub>x</sub>, organics, and possibly oxygen; and extensive gas pretreatment is required for a chemical plant application. Page 5/8

The fluorocarbon absorption system effectively removes krypton and xenon in the concentration ranges typical of those characteristic of dissolver or vessel off-gas streams; the system can tolerate CO<sub>2</sub> and nitrous oxides in the feed gas at concentrations approaching 1% volume. The krypton-xenon product is concentrated and purified in a fractionation and stripping column, and subsequently bottled for permanent storage or disposal.

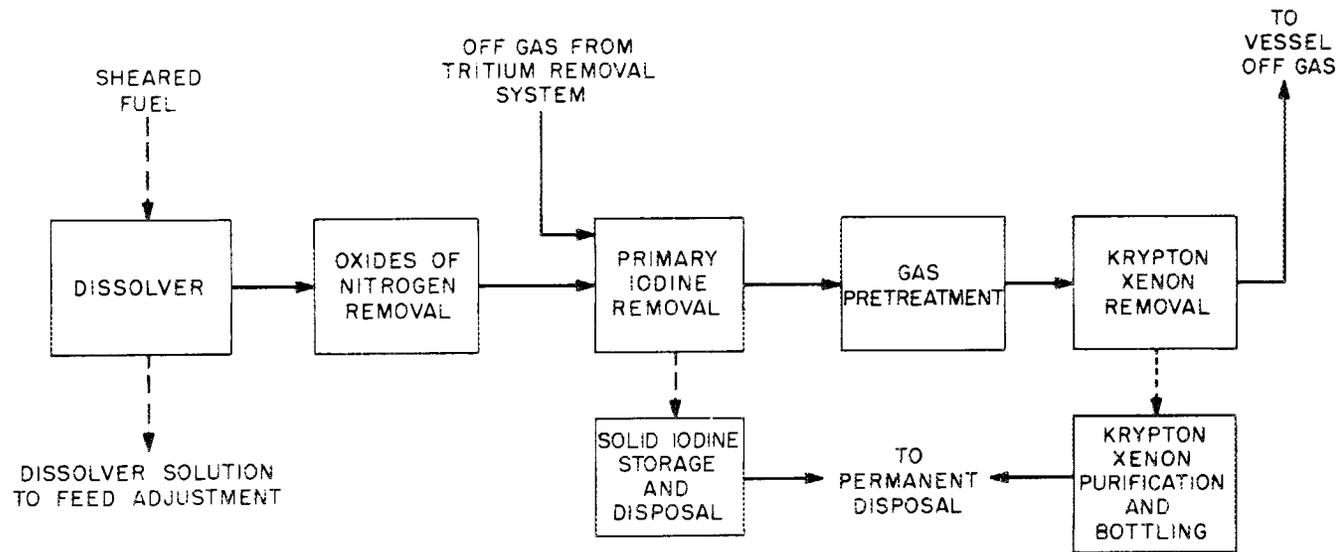


Fig. 4.2. Flowsheet for Krypton and Primary Iodine Removal.

Treatment of the total plant off-gas for krypton removal is practical if the total off-gas flow rate is kept low (100 cfm); and such an approach can be used either instead of or in addition to primary control. However, if krypton is removed only in the secondary off-gas treatment system, excessively high krypton concentrations in the recycle cell and process gases may result. For overall krypton retention factors of  $10^3$  or less, primary retention is recommended; for significantly higher retention, combined primary and secondary treatment is required when using the proposed flow arrangement.

#### 4.4 Iodine Control

Effective control of iodine in a fuel reprocessing plant depends on an understanding of iodine behavior in each of the process steps, as well as a demonstrated capability for effective removal of iodine from liquid and gaseous process streams. The iodine control system proposed for "near zero" release fuel reprocessing plants includes five major areas of treatment, including the following:

- (1) Evolution of iodine from the dissolver solution.
- (2) Primary iodine removal from head-end off-gas.
- (3) Secondary iodine removal from total cell and equipment off-gas.
- (4) Final iodine removal from plant gaseous effluents.
- (5) Iodine removal from recycle process water and acid.

The application of these systems to the overall retention of iodine is indicated in Fig. 3.1 and in much more detail in the Appendix (Figs. A-4 through A-9).

Evolution of iodine from the dissolver solution to a level of  $< 1\%$  of that initially present in the fuel is desirable to prevent the bulk ( $\sim 2.5$  kg of iodine per day for a 5-ton/day plant) of the iodine from entering the solvent extraction complex. This not only reduces the load on secondary iodine removal equipment but also minimizes the formation of difficultly trapped organic iodides. Evolution of 95 to 98% of the iodine can be accomplished relatively easily by steam-stripping of the

dissolver solution in the presence of excess  $\text{NO}_x$ . Further removal requires the addition of stable iodine carrier followed by treatment to obtain isotopic exchange between the residual activity and added carrier. Further evolution of iodine is then obtained by additional steam-stripping.

All off-gas streams from the head-end operations are combined and fed to the primary iodine removal system, which should be designed to give an iodine decontamination factor in the range of  $10^4$ . The primary iodine removal system should have the capability for handling a few kilograms of iodine per day and should recover the iodine in a form amenable to permanent disposal. Of two aqueous scrubbing systems under development, the IODEX system, using 20 molar nitric acid, appears to be the best choice. The alternate system, which employs 8-10 molar nitric acid and 0.2-0.4 molar mercuric nitrate, is sensitive to contaminants in the feed gas stream. Both systems oxidize iodine forms to higher nonvolatile species and deliver a solid waste composed of iodates.

Iodine that bypasses the primary iodine removal system distributes throughout the downstream process steps and eventually appears in the process off-gas and recycle acid and water streams. The total cell and equipment off-gas is passed through a secondary iodine removal system, where an iodine DF in excess of  $10^4$  is desired. The secondary off-gas treatment also includes steps for removing particulates, ruthenium, and chemical constituents such as nitrous oxides. Most of the gaseous effluents from secondary treatment are recycled for process use. Any excess is routed to the final treatment system prior to discharge to the environment. The aqueous scrubber systems previously described, as well as solid sorbents such as silver zeolite, are applicable for use in secondary iodine removal.

Final off-gas treatment includes an iodine removal system having a decontamination capability in excess of  $10^4$ , giving an overall plant retention factor of  $>10^{10}$  for iodine. A solid sorbent such as silver zeolite appears to be the logical choice for the final iodine removal system. Off-gas flowing through the final treatment system will include the total air inleakage to the cell bank plus any net process air or gas added from outside. Ideally, this total flow rate is less than 100 cfm.

A significant fraction of the iodine reaching the process equipment downstream of the dissolver will follow the aqueous stream and terminate in the recovered water and acid. Iodine can be evolved from water and dilute acid ( $< 4 M$ ) by steam-stripping and other techniques that are discussed under item (1) given on page 23. Trace concentrations of iodine can be removed from water and dilute ( $< 4 M$ ) acid by adsorption on activated charcoal or organic resins. Essentially complete removal of iodine from recycle water and acid streams that are to be used in the final stage of off-gas treatment is necessary to prevent recontamination of relatively iodine-free gas streams.

#### 4.5 Recovery and Recycle of Acid and Water

The ideal of "near zero" release in fuel reprocessing can be more closely approached when the release of water, in any form, to the environment is essentially zero. This requires that there not be any discharge of water and that all plant off-gas be dried to a very low dewpoint. This approach is mandatory if the overall plant retention factor for tritium is to be derived totally or partially by retention of plant water. If tritium retention is not required or is obtained by other means, the discharge of small quantities of water vapor, after complete decontamination from all activities except tritium, is an acceptable alternative.

Two problems that must be successfully resolved in order to attain total water and acid recycle are: (1) minimization of the net input of water and acid, and (2) purification of recycle acid and water to a degree that will prevent process interference. Input of water and acid, as such, can be held to very minimal quantities by recycle; however, a number of other sources of water must be considered. These sources of water are summarized in Table 4.1. Little can be done about the humidity of inleaking air except to minimize inleakage. Addition of hydrogen can be avoided by in-cell electrolysis of water. If organic materials are to be disposed of in-cell by incineration, a net water input will be realized. A typical water input of 60 kg/day corresponds to an accumulated inventory of 175,000 gal over the 30-year plant life. Process use of  $\text{NO}_x$  could be equivalent to the addition of 100 to 300 kg of  $\text{HNO}_3$  per day.

This net addition can be avoided by recovering and recycling  $\text{NO}_x$  produced in the denitration of  $\text{UO}_2(\text{NO}_3)_2$  or in the waste solidification process.

Table 4.1. Sources of Water Input to Reprocessing Plant  
(Basis: 5 tons/day U + Pu charged)

	Input Range (kg/day)	Typical Value (kg/day)
Humidity of air inleakage to cell	3-300	35 <sup>a</sup>
Hydrogen added to reduce $\text{UO}_3$ to $\text{UO}_2$	0-300	0 <sup>b</sup>
Process chemical addition	5-50	10
Incineration of organic materials	0-50	15
Total	8-700	60

<sup>a</sup>Assumes an air inleakage of 100 cfm at 20°C and 50% relative humidity.

<sup>b</sup>In-cell production of hydrogen by electrolysis of recycle water.

The second problem associated with recycle is the purification of the recycle water and acid from both radioactive and chemical constituents (such as dissolved organics) that might interfere with process operations. Particularly difficult active species include iodine and ruthenium. A second stage of purification may be required for recycle streams that are used for product stripping and handling or for final off-gas treatment steps.

## 5. MODELING AND COMPUTER ANALYSIS

The behavior of important chemical and fission product components in each of 24 process steps (Fig. A-1 in the Appendix) was modeled, and a computer program was written to calculate the distributions of each

component throughout the process. Internal modeling of individual systems was kept simple, with the primary emphasis being placed on the effect of interaction between systems and the effect of recycle on overall containment. Variables that were studied relative to their effect on overall plant containment include the following:

- (1) Air inleakage rate to cells.
- (2) Recycle air rates to cells.
- (3) Off-gas routing and treatment options.
- (4) Degree of water removal from off-gas streams.
- (5) Fraction of fission products escaping to the atmosphere of head-end cells.
- (6) Fraction of iodine evolved in feed preparation steps.
- (7) Effectiveness of specific effluent treatment systems.

The main emphasis of this study was centered on effluent treatment; none of the operating characteristics of the main-line process was varied unless it was suspected to have a direct effect on effluent control. The effects of the above variables on the release rates and distribution of important fission products are discussed in the following section.

Table 5.1 summarizes the seven basic air flow patterns that were used in all runs. Outside air is assumed to leak into each of eight process cells at rates varying from 0.5 to 100 scfm, as indicated in the table. Additional gas is added for process use at the rate of 0.9 cfm. Off-gas from the head-end cells, including receiving and storage, mechanical handling, and dissolution and feed preparation, may be routed either to the dissolver off-gas or to the vessel off-gas, and may thus bypass the primary systems. The fraction of off-gas routed to the vessel off-gas in each flow option is indicated. Approximate flow rates through important internal treatment systems are listed; and the distribution of treated vessel off-gas as recycle air to cells, recycle process air, or net off-gas to final treatment is tabulated.

Other process options used in individual runs will be described in detail in later sections.

Table 5.1. Basic Air Flow Patterns for Case Studies

	Air Flow Pattern						
	1	2	3	4	5	6	7
Input Stream, scfm							
Outside air inleakage to cells, scfm							
Analytical and final off-gas cells (2)	2	2	2	20	20	20	20
Kr-Xe removal equipment cell (1)	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Other process cells (5)	5	5	5	50	50	50	500
Total air inleakage	7.5	7.5	7.5	70.5	70.5	70.5	520.5
Outside addition of process gas, scfm	0.9	0.9	0.9	0.9	0.9	0.9	0.9
Internal Routing							
Fraction of cell off gas to secondary treatment							
Receiving and storage cell	0	0	0	0	0	0	1.0
Mechanical handling cell	0	0.9	0	0.9	0.99	0.90	0.99
Dissolver and feed preparation cell	0	0.9	0	0.9	0.99	0.90	0.99
Internal Flow Rates, scfm							
Primary tritium system effluent	1.0	1.1	11.0	1.0	1.1	11.0	1.0
Primary iodine system effluent	2.8	3.0	22.8	11.8	12.0	31.8	1.8
Primary Kr-Xe system effluent	3.3	3.5	23.3	12.3	12.5	32.2	2.3
Secondary iodine system effluent	14.3	71.2	64.2	77.3	677	667	528
Vessel off-gas treatment system effluent	14.8	74.7	69.7	77.8	677.7	672.7	528.3
Recycle process air	6.9	6.9	6.9	6.9	6.9	6.9	6.9
Recycle cell air	0	60	60	0	600	600	0
To final off-gas treatment	7.9	7.8	2.8	70.9	70.8	65.8	521.4
Output Streams, scfm							
Final off-gas treatment effluent to stack	8.4	8.4	8.3	71.2	71.2	71.2	521.4
Kr-Xe waste gas	0.02	0.02	0.06	0.16	0.16	0.2	0.01

### 5.1 Distribution and Removal of Tritium

The behavior of tritium in the process system was investigated in a series of 12 computer runs. The conditions for these runs are listed in Table 5.2, and the results are summarized in Table 5.3. Runs 1-7 illustrate the effects of varying off-gas rates and routing on tritium distribution. Runs 8 and 10 illustrate the effect of higher tritium release to the storage pool and head-end cell atmosphere, while runs 13 and 14 illustrate the effect of omitting the voloxidation and primary tritium removal system. In all runs except run 4A, a secondary Kr-Xe removal system was assumed to operate on the plant off-gas prior to discharge to the atmosphere and a tritium DF of 200 was assigned to this system. If this final Kr-Xe removal system was omitted, the overall plant DF for tritium would be decreased by a factor of 200 for all cases except 4A.

The amount of tritium in the net off-gas stream is controlled by the water content of the off-gas and the tritium inventory in the plant. In runs 1-6 the net plant off-gas was dried by means of a zeolite bed to a  $-140^{\circ}\text{F}$  dewpoint, yielding overall plant retention factors ranging from  $5.6 \times 10^9$  to  $5.4 \times 10^{10}$ . In general, the DF was inversely proportional to off-gas rates and was affected significantly by the bypass of tritium around the head-end treatment system (compare runs 2 and 3). Recycle of cell air had little effect on overall tritium removal. In run 4A (as compared to 4), the final Kr-Xe removal system was not operated and the effect was a reduction in the overall tritium DF by a factor of 200. In run 7 the off-gas rate was increased to 520 cfm and a final off-gas dewpoint of  $32^{\circ}\text{F}$  was assumed, resulting in an overall tritium DF of only 260.

The effect of a higher head-end release on the distribution of tritium for two air flow patterns is shown in runs 8 and 10. The fraction of tritium released to the receiving and storage pool and to the mechanical handling off-gas was doubled as compared with that in runs 5 and 7. The amount of tritium in the recycle acid and water systems increased by a factor of 1.6 in each case. This resulted in a proportional increase in the tritium released to the two off-gas streams.

Table 5.2. Run Conditions for Tritium Behavior Study

Run No.	Air Flow Pattern <sup>a</sup>	Head-End Tritium Release (Fraction of Input)		Secondary Krypton System <sup>b,c</sup>	Voloxidizer and Primary Tritium Removal <sup>d</sup>
		Storage Pool	Mechanical Cell		
1	1	0.005	0.005	Yes	Yes
2	2	0.005	0.005	Yes	Yes
3	3	0.005	0.005	Yes	Yes
4	4	0.005	0.005	Yes	Yes
4A	4	0.005	0.005	No	Yes
5	5	0.005	0.005	Yes	Yes
6	6	0.005	0.005	Yes	Yes
7	7	.005	.005	Yes	Yes
8	5	.01	.01	Yes	Yes
10	7	.01	.01	Yes	Yes
13	5	.005	.005	Yes	No
14	7	.005	.005	Yes	No

<sup>a</sup>See Table 4.1.

<sup>b</sup>Tritium DF of ~200 assumed across Kr-Xe system.

<sup>c</sup>This system is optional.

<sup>d</sup>98% evolution of tritium from fuel in voloxidizer with -140°F dewpoint in exit from primary tritium system.

Table 5.3. Distribution of Tritium in Process Streams

Run No.	Tritium Content of Plant Effluent Streams (Fraction of $^3\text{H}$ Input)					Tritium Content of Internal Process Streams (Fraction of $^3\text{H}$ Input)			Overall Plant Tritium DF
	Total Off Gas	Excess Liquid to Storage	Tritium Waste Stream	Solid Waste	U and Pu Products	Effluent from Primary $^3\text{H}$ Removal	Bypass Primary Tritium System	Recycle Water and Acid	
1	$1.9 \times 10^{-11}$	$3.46 \times 10^{-3}$	0.9899	$6.57 \times 10^{-3}$	$1.30 \times 10^{-7}$	$1.9 \times 10^{-5}$	$3.6 \times 10^{-5}$	4.45	$5.3 \times 10^{10}$
2	$3.5 \times 10^{-11}$	$5.7 \times 10^{-3}$	0.9899	$6.57 \times 10^{-3}$	$1.30 \times 10^{-7}$	$2.4 \times 10^{-6}$	$6.3 \times 10^{-3}$	7.54	$2.8 \times 10^{10}$
3	$1.8 \times 10^{-11}$	$3.6 \times 10^{-3}$	0.9899	$6.57 \times 10^{-3}$	$1.30 \times 10^{-7}$	$1.9 \times 10^{-4}$	$3.6 \times 10^{-5}$	4.74	$5.4 \times 10^{10}$
4	$1.7 \times 10^{-9}$	$5.7 \times 10^{-3}$	0.9899	$6.57 \times 10^{-3}$	$1.30 \times 10^{-7}$	$1.9 \times 10^{-5}$	$6.7 \times 10^{-3}$	7.66	$5.8 \times 10^9$
4A	$3.5 \times 10^{-7}$	$5.6 \times 10^{-3}$	0.9899	$6.57 \times 10^{-3}$	$1.30 \times 10^{-7}$	$1.9 \times 10^{-5}$	$6.7 \times 10^{-3}$	7.66	$2.9 \times 10^6$
5	$1.8 \times 10^{-9}$	$6.3 \times 10^{-3}$	0.9831	$1.06 \times 10^{-2}$	$2.07 \times 10^{-7}$	$2.2 \times 10^{-5}$	$9.3 \times 10^{-3}$	10.7	$5.6 \times 10^9$
6	$1.7 \times 10^{-9}$	$5.88 \times 10^{-3}$	0.9838	$1.03 \times 10^{-2}$	$2.01 \times 10^{-7}$	$1.1 \times 10^{-4}$	$8.4 \times 10^{-3}$	10.4	$5.9 \times 10^9$
7	$3.8 \times 10^{-3}$	$9.1 \times 10^{-3}$	0.9838	$1.03 \times 10^{-2}$	$2.01 \times 10^{-7}$	$1.9 \times 10^{-5}$	$9.3 \times 10^{-3}$	3.49	260
8	$2.8 \times 10^{-9}$	$9.89 \times 10^{-3}$	0.9734	$1.67 \times 10^{-2}$	$3.29 \times 10^{-7}$	$2.2 \times 10^{-5}$	$1.6 \times 10^{-2}$	16.8	$3.5 \times 10^9$
10	$6.1 \times 10^{-3}$	$1.43 \times 10^{-2}$	0.9733	$6.30 \times 10^{-3}$	$1.14 \times 10^{-7}$	$1.8 \times 10^{-5}$	$1.6 \times 10^{-2}$	5.58	165
13	$1.010^{-7}$	0.382	0	$6.30 \times 10^{-3}$	$1.14 \times 10^{-7}$	-	1.00	627	$9.5 \times 10^8$
14	0.226	0.541	0	0.233	$4.00 \times 10^{-5}$	-	1.00	208	4.4

In runs 13 and 14, the standard release rates were used while the voloxidation and primary tritium removal systems were deactivated. The amounts of tritium in the recycle acid and water systems increased by a factor of 59. In run 13, where the net off-gas is dried in a zeolite bed, good retentions of tritium were still obtained (DF,  $\sim 9.5 \times 10^6$ ). However, in the case where the off-gas is not passed through a zeolite bed (run 14), the DF fell to 4.4. In both of these runs, the excess liquid stream contains a large fraction of the tritium input: 38% in run 5, and 54% in run 14.

In conclusion, operation of the final off-gas treatment has the controlling influence on the amount of tritium being released in the stack gas. The degree of drying is the major factor since the tritium is in the form of tritiated water. The secondary krypton removal system also removes water from the dried gas stream and hence will also remove a proportional amount of the remaining tritium. Voloxidation and tritium removal can be used to obtain an overall DF of approximately 100 without water retention. With low off-gas rates, zeolite beds can be used to dry the total off-gas; and, under these conditions, the voloxidation and tritium removal steps are not as critical since DFs of the order of  $10^6$  can be obtained by drying alone. In this case the plant system must tolerate the large tritium inventory in recycle water. Voloxidation, tritium removal, and extensive drying of the off-gas are required in order to obtain DFs in the  $10^8$ - $10^{10}$  range. Although this degree of treatment could be applied, such extensive measures greatly exceed the practical.

## 5.2 Distribution and Removal of Krypton

Some krypton is assumed to escape from the fuel elements during handling in the storage cell (0.1%) and the mechanical handling cell (0.5%). The remainder is evolved in the shear, the voloxidizer, and the dissolver. A primary krypton removal system with a system DF of  $\sim 1000$  is provided for the dissolver off-gas stream and is located downstream of the primary iodine removal equipment. A secondary krypton removal system is included as part of the final off-gas treatment (system O) and is

assumed to give a system DF of 1000. The distribution of krypton in plant off-gas streams is summarized in Table 5.4. The effectiveness of the primary removal system, which is determined by the fraction of krypton that bypasses the system, is in the range of  $10^3$  in cases where there is no intentional bypass of head-end cell off-gas (runs 1 and 3). For cases where there is significant bypass of cell air around the primary removal system, the effective DF across the primary system is of the order of 100 to 200.

Two additional calculations were made to demonstrate the effect of having no primary krypton removal system while retaining the secondary system. Run 5P used the same air flow pattern as run 5, and run 7P used the same air flow pattern as run 7. Both of these runs had identical DFs of  $\sim 1000$ , the DF of the secondary krypton removal system. All of the krypton in the plant feed reports to the secondary krypton removal system. In run 5P, where a large amount of recycle cell air is used, high concentrations of krypton are present in the off-gas system. If neither of the krypton removal systems were operated, the distribution of krypton in the off-gas streams would be similar to that shown in runs 5P and 7P, with the exception that all the krypton fed to the plant would be released through the stack.

An additional series of runs was made to investigate the effects of increased head-end release and the omission of voloxidation on krypton distribution. In runs 8 and 9, the amount of krypton released to the receiving and storage cell atmosphere was increased by a factor of 10, while the amount of krypton released to the mechanical handling cell was increased by a factor of 2. The overall krypton retention factor decreased by factors of 1.7 (comparing run 8 with run 5) and 3.1 (comparing run 9 with run 5A).

In run 9 the off-gas from the receiving and storage cell was routed to the vessel off-gas, causing a significant increase in the amount of krypton that bypassed the primary krypton system; the secondary krypton removal system was not operated.

Table 5.4. Distribution of Krypton in Process Off-Gas

Run No.	Air Flow Pattern	Fraction of Input						
		Inlet to Vessel Off-Gas Treatment	Head-End <sup>b</sup> Off-Gas Bypassing Primary Kr Treatment	Outlet from Primary Kr Treatment	Recycle Air	Feed to Final Kr Removal	Discharge to Stack	Plant Retention Factor
1	1	$2.2 \times 10^{-3}$	0.0	$1.01 \times 10^{-3}$	0.001	$1.2 \times 10^{-3}$	$1.2 \times 10^{-6}$	$8.3 \times 10^5$
2	2	$5.0 \times 10^{-2}$	0.018	$1.00 \times 10^{-3}$	0.045	$5.6 \times 10^{-3}$	$5.6 \times 10^{-6}$	$1.8 \times 10^5$
3	3	$2.5 \times 10^{-3}$	0.0	$1.01 \times 10^{-3}$	0.0025	$3.0 \times 10^{-4}$	$3.0 \times 10^{-7}$	$3.3 \times 10^6$
4	4	$7.5 \times 10^{-3}$	0.0057	$1.00 \times 10^{-3}$	$6.6 \times 10^{-4}$	$6.8 \times 10^{-3}$	$6.9 \times 10^{-6}$	$1.5 \times 10^5$
4A	4	$7.5 \times 10^{-3}$	0.0057	$1.00 \times 10^{-3}$	$6.6 \times 10^{-4}$	$6.8 \times 10^{-3}$	$6.8 \times 10^{-3}$	150
5	5	$6.8 \times 10^{-2}$	0.026	$1.00 \times 10^{-3}$	0.061	$7.2 \times 10^{-3}$	$7.3 \times 10^{-6}$	$1.4 \times 10^5$
5A	5	$6.8 \times 10^{-2}$	0.026	$1.00 \times 10^{-3}$	0.061	$7.1 \times 10^{-3}$	$7.2 \times 10^{-3}$	140
5P	5	9.50	2.78	1.02	8.51	1.00	$1.0 \times 10^{-3}$	1000
6	6	$5.0 \times 10^{-2}$	0.019	$1.00 \times 10^{-3}$	0.045	$5.3 \times 10^{-3}$	$5.4 \times 10^{-6}$	$1.9 \times 10^5$
7	7	$8.5 \times 10^{-3}$	0.0073	$1.00 \times 10^{-3}$	$1.1 \times 10^{-4}$	$8.3 \times 10^{-3}$	$8.3 \times 10^{-3}$	120
7P	7	1.01	0.0074	0.993	0.013	1.00	$1.0 \times 10^{-3}$	1000
8	5	$1.1 \times 10^{-1}$	0.042 <sup>b</sup>	$1.00 \times 10^{-3}$	0.101	$1.2 \times 10^{-2}$	$1.2 \times 10^{-6}$	$8.3 \times 10^4$
9	5	$2.1 \times 10^{-1}$	0.081 <sup>b</sup>	$9.90 \times 10^{-4}$	0.184	$2.1 \times 10^{-2}$	$2.2 \times 10^{-2}$	45
13	5	$6.8 \times 10^{-2}$	0.026	$1.00 \times 10^{-3}$	0.061	$7.1 \times 10^{-3}$	$7.2 \times 10^{-6}$	$1.4 \times 10^5$

(a) Krypton releases of 0.1% to the storage cell and 0.5% to mechanical handling cell are assumed.

(b) Krypton releases of 1% to both storage and mechanical handling cells are assumed for runs 8 and 9.

Several general conclusions can be drawn from the krypton data for the runs just discussed. In order to maximize the amount of krypton retained, the amount of krypton bypassing the primary system should be minimized. This can be accomplished in either of two ways. If all the cell effluent air is taken into the head-end equipment, the amount of krypton released in these cells is immaterial since all of it would be sent to the primary krypton removal system in the various equipment off-gas streams. However, if a portion of the cell atmosphere bypasses primary treatment, then the head-end releases should be minimized in order to minimize the effect of bypassing the primary system. The use of recycle air to sweep the cells is beneficial, especially if a significant proportion of the cell effluent gas is taken into the equipment off-gas. The recycle air, in effect, returns any krypton which either bypasses or exits from the primary system for further processing.

Primary krypton removal will provide a DF of 1000 if all the head-end gas is passed through the system. With most of the runs, a certain amount of the head-end off-gas bypasses the primary krypton system and DFs in the range of 50-150 were typical. Operation of the secondary system alone will also produce a DF of approximately 1000. However, the plant must be able to tolerate the high krypton content of the cell gas that occurs with large recycle air rates. To obtain DFs in excess of  $10^2$ - $10^3$ , both the primary and secondary systems were required. The installation of both systems has the advantage that plant operation could probably be continued in the event of a shutdown of either of the systems. If only one krypton removal system were installed, a failure in that system would require the entire plant to be shut down or would result in all the krypton being released to the atmosphere.

### 5.3 Distribution and Retention of Iodine

The complexity of iodine behavior and the many possible interactions between process systems make a simple analysis of iodine distribution impractical. Two radioactive isotopes of iodine, namely  $^{131}\text{I}$  (half-life, 8 days) and  $^{129}\text{I}$  (half-life,  $1.6 \times 10^7$  years) may be present in the fuel at the time of processing. The retention of  $^{131}\text{I}$  will, in general,

be equal to or greater than that of  $^{129}\text{I}$  due to some gain from decay of  $^{131}\text{I}$  in the process inventory. For this study,  $^{129}\text{I}$  was taken as the isotope to follow and no credit was taken for radioactive decay.

A series of computer runs (described in Table 5.5) was made to illustrate the distribution of iodine throughout the process system for a set of basic air flow patterns and typical operating conditions. The distribution of iodine in plant gas streams is summarized in Table 5.6; the distribution in solid and liquid streams is summarized in Table 5.7. The actual system DF (Table 5.8) is defined as the ratio of the iodine content of the inlet stream to that of the outlet stream and the effective DF is defined as the ratio of the iodine content in the inlet to a system to that at the next downstream iodine system inlet. (See Fig. A-1 in the Appendix for an overall plant flowsheet with identifying stream codes.)

Runs 1-7 illustrate the iodine distribution for the basic flow patterns given in Table 5.1. Overall plant retention factors in the range of  $10^{11}$  to  $10^{12}$  were typical, with no great difference being produced by internal air flow patterns or gas flow rates. The limiting overall DF was, in most cases, set by recontamination of the off-gas in the final stage of treatment with recycle water or the bypass of small quantities of iodine around the final treatment system. As indicated in Table 5.8, the effective DF of the final off-gas treatment system was considerably less than the actual DF in some of the runs. Runs 6, 6A, and 7 represent runs in which the input to final off-gas treatment was higher by two to three orders of magnitude than previous runs, yet the final removal system was effective in giving further decontamination.

The effectiveness of the vessel off-gas treatment system varies widely and depends on such factors as system efficiency, degree of recontamination, and the ratio of treated off-gas recycled to the cell to that going to final off-gas treatment. For example, in run 5 (Table 5.7), the actual vessel off-gas iodine DF was only 1.0, while the effective DF was  $\sim 10^4$ . This effect was the result of processing the air going to the final off-gas stream through efficient iodine removal equipment while bypassing the gas being recycled to the cell around the iodine removal system. While such options may influence the general level of iodine contamination in the cells, they have little effect on overall iodine retention.

Table 5.5. Run Conditions for Basic Iodine Behavior Study

Run	Air Flow	Head-End Fractional Release			Fraction of I <sub>2</sub> Evolved from Dissolver Solution
		Receiving and Storage Water	Receiving and Storage Air	Mechanical Handling	
1	1	0.005	0.001	0.005	0.999
2	2	0.005	0.001	0.005	0.999
3	3	0.005	0.001	0.005	0.999
4	4	0.005	0.001	0.005	0.999
4A <sup>a</sup>	4	0.005	0.001	0.005	0.999
5	5	0.005	0.001	0.005	0.999
5A <sup>a</sup>	5	0.005	0.001	0.005	0.999
6	6	0.005	0.001	0.005	0.999
6A <sup>a</sup>	6	0.005	0.001	0.005	0.999
7 <sup>a</sup>	7 <sup>b</sup>	0.005	0.001	0.005	0.999
10 <sup>a</sup>	7	0.01	0.01	0.01	0.999
11	3	0.01	0.01	0.01	0.999
14 <sup>a</sup>	7	0.005	0.001	0.005	0.999
15	1	0.005	0.001	0.005	0.99
18 <sup>a</sup>	7	0.005	0.001	0.005	0.99
19	1	0.005	0.001	0.005	0.95
22 <sup>a</sup>	7	0.005	0.001	0.005	0.95
23 <sup>a</sup>	7	0.01	0.01	0.01	0.95

<sup>a</sup>Final krypton system was not operated. This system was assumed to give a DF of ~20 for I<sub>2</sub>.

<sup>b</sup>No voloxidation.

Table 5.6. Distribution of Iodine in Plant Gas Streams<sup>a</sup>  
(Fraction of Input)

Run	Gaseous Bypass Primary I <sub>2</sub> Removal, A2 + B3	Feed Primary I <sub>2</sub> Removal, F1	Off-Gas Primary I <sub>2</sub> Removal, F1	Off-Gas Secondary I <sub>2</sub> Removal, M6	I <sub>2</sub> Removal, M1	Recycle Air N2 + N3	Feed Final Off-Gas Treatment, N1	Off-Gas Final O-G Treatment, O1
1	0.0	0.9771	4.92 x 10 <sup>-8</sup>	5.12 x 10 <sup>-3</sup>	3.63 x 10 <sup>-7</sup>	2.21 x 10 <sup>-10</sup>	2.53 x 10 <sup>-10</sup>	7.67 x 10 <sup>-13</sup>
2	5.01 x 10 <sup>-3</sup>	0.9742	4.92 x 10 <sup>-8</sup>	1.03 x 10 <sup>-2</sup>	7.32 x 10 <sup>-7</sup>	6.82 x 10 <sup>-10</sup>	7.98 x 10 <sup>-11</sup>	1.22 x 10 <sup>-12</sup>
3	0.0	0.9792	4.92 x 10 <sup>-8</sup>	5.12 x 10 <sup>-3</sup>	3.66 x 10 <sup>-7</sup>	4.59 x 10 <sup>-10</sup>	1.95 x 10 <sup>-11</sup>	7.67 x 10 <sup>-13</sup>
4	5.01 x 10 <sup>-3</sup>	0.9742	4.92 x 10 <sup>-8</sup>	1.03 x 10 <sup>-2</sup>	7.32 x 10 <sup>-7</sup>	6.74 x 10 <sup>-11</sup>	6.94 x 10 <sup>-10</sup>	1.30 x 10 <sup>-12</sup>
4A	5.01 x 10 <sup>-3</sup>	0.9742	4.92 x 10 <sup>-8</sup>	1.03 x 10 <sup>-2</sup>	7.32 x 10 <sup>-7</sup>	6.73 x 10 <sup>-11</sup>	6.94 x 10 <sup>-10</sup>	1.45 x 10 <sup>-12</sup>
5	5.51 x 10 <sup>-3</sup>	0.9737	4.92 x 10 <sup>-8</sup>	1.08 x 10 <sup>-2</sup>	8.37 x 10 <sup>-7</sup>	8.07 x 10 <sup>-7</sup>	8.90 x 10 <sup>-11</sup>	5.09 x 10 <sup>-12</sup>
5A	5.51 x 10 <sup>-3</sup>	0.9737	4.92 x 10 <sup>-8</sup>	1.08 x 10 <sup>-2</sup>	8.37 x 10 <sup>-7</sup>	8.07 x 10 <sup>-7</sup>	8.90 x 10 <sup>-11</sup>	3.06 x 10 <sup>-12</sup>
6	5.01 x 10 <sup>-3</sup>	0.9742	4.92 x 10 <sup>-8</sup>	1.03 x 10 <sup>-2</sup>	7.99 x 10 <sup>-7</sup>	7.68 x 10 <sup>-7</sup>	8.43 x 10 <sup>-8</sup>	3.06 x 10 <sup>-12</sup>
6A	5.01 x 10 <sup>-3</sup>	0.9742	4.92 x 10 <sup>-8</sup>	1.03 x 10 <sup>-2</sup>	7.99 x 10 <sup>-7</sup>	7.68 x 10 <sup>-7</sup>	8.43 x 10 <sup>-8</sup>	7.60 x 10 <sup>-11</sup>
7	6.51 x 10 <sup>-3</sup>	0.9727	4.92 x 10 <sup>-8</sup>	1.19 x 10 <sup>-2</sup>	8.78 x 10 <sup>-7</sup>	1.10 x 10 <sup>-11</sup>	9.35 x 10 <sup>-7</sup>	7.78 x 10 <sup>-10</sup>
10	2.03 x 10 <sup>-2</sup>	0.9542	4.91 x 10 <sup>-8</sup>	3.02 x 10 <sup>-2</sup>	2.21 x 10 <sup>-6</sup>	2.65 x 10 <sup>-11</sup>	2.23 x 10 <sup>-6</sup>	1.85 x 10 <sup>-9</sup>
11	0.0	0.9742	4.91 x 10 <sup>-8</sup>	9.65 x 10 <sup>-3</sup>	6.90 x 10 <sup>-7</sup>	8.87 x 10 <sup>-10</sup>	3.76 x 10 <sup>-11</sup>	1.47 x 10 <sup>-12</sup>
14	6.51 x 10 <sup>-3</sup>	0.9878	4.99 x 10 <sup>-8</sup>	1.19 x 10 <sup>-2</sup>	8.78 x 10 <sup>-7</sup>	1.10 x 10 <sup>-11</sup>	9.35 x 10 <sup>-7</sup>	7.76 x 10 <sup>-10</sup>
15	0.0	0.9735	4.91 x 10 <sup>-8</sup>	1.02 x 10 <sup>-2</sup>	7.20 x 10 <sup>-7</sup>	3.54 x 10 <sup>-10</sup>	4.06 x 10 <sup>-10</sup>	1.21 x 10 <sup>-12</sup>
18	6.51 x 10 <sup>-3</sup>	0.9671	4.91 x 10 <sup>-8</sup>	1.70 x 10 <sup>-2</sup>	1.24 x 10 <sup>-6</sup>	1.47 x 10 <sup>-11</sup>	1.24 x 10 <sup>-6</sup>	1.03 x 10 <sup>-9</sup>
19	0.0	0.9486	4.89 x 10 <sup>-8</sup>	3.28 x 10 <sup>-2</sup>	2.30 x 10 <sup>-6</sup>	9.47 x 10 <sup>-10</sup>	1.08 x 10 <sup>-9</sup>	3.23 x 10 <sup>-12</sup>
22	6.51 x 10 <sup>-3</sup>	0.9422	4.89 x 10 <sup>-8</sup>	3.96 x 10 <sup>-2</sup>	2.83 x 10 <sup>-6</sup>	3.13 x 10 <sup>-11</sup>	2.59 x 10 <sup>-6</sup>	2.14 x 10 <sup>-9</sup>
23	2.03 x 10 <sup>-2</sup>	0.9243	4.88 x 10 <sup>-8</sup>	5.74 x 10 <sup>-2</sup>	4.13 x 10 <sup>-6</sup>	4.65 x 10 <sup>-11</sup>	3.86 x 10 <sup>-6</sup>	3.18 x 10 <sup>-9</sup>

<sup>a</sup>Designation of streams as shown in Fig. A-1 in the Appendix.

Table 5.7. Distribution of Iodine in Solid and Liquid Streams<sup>a</sup>  
(Fraction of Input)

Run No.	Slurried Resin to Solid Waste Handling, A4	Pool H <sub>2</sub> O to Acid-H <sub>2</sub> O Recovery, A5	Feed to Solvent Extraction, DE1	Recycle Water and Acid	Tritium Waste Stream, W6A	Misc. Solid Waste, W5	Iodine Solid Waste, W6B
1	$2.5 \times 10^{-4}$	$4.7 \times 10^{-3}$	$6.2 \times 10^{-4}$	$3.0 \times 10^{-6}$	$1.5 \times 10^{-2}$	$1.2 \times 10^{-2}$	0.984
3	$2.5 \times 10^{-4}$	$4.7 \times 10^{-3}$	$6.2 \times 10^{-4}$	$3.0 \times 10^{-6}$	$1.5 \times 10^{-2}$	$1.2 \times 10^{-2}$	0.984
7	$2.5 \times 10^{-4}$	$4.7 \times 10^{-3}$	$6.2 \times 10^{-4}$	$3.0 \times 10^{-6}$	$1.5 \times 10^{-2}$	$1.2 \times 10^{-3}$	0.984
10	$5.0 \times 10^{-4}$	$9.5 \times 10^{-3}$	$6.1 \times 10^{-4}$	$5.6 \times 10^{-6}$	$1.5 \times 10^{-2}$	$1.6 \times 10^{-3}$	0.984
11	$5.0 \times 10^{-4}$	$9.5 \times 10^{-3}$	$6.1 \times 10^{-4}$	$5.6 \times 10^{-6}$	$1.5 \times 10^{-2}$	$1.6 \times 10^{-3}$	0.984
15	$2.5 \times 10^{-4}$	$4.7 \times 10^{-3}$	$6.2 \times 10^{-3}$	$6.0 \times 10^{-6}$	$1.5 \times 10^{-2}$	$1.7 \times 10^{-3}$	0.983
18	$2.5 \times 10^{-4}$	$4.7 \times 10^{-3}$	$6.2 \times 10^{-3}$	$6.0 \times 10^{-6}$	$1.5 \times 10^{-2}$	$1.7 \times 10^{-3}$	0.984
19	$2.5 \times 10^{-4}$	$4.7 \times 10^{-3}$	$3.1 \times 10^{-2}$	$1.9 \times 10^{-5}$	$1.5 \times 10^{-2}$	$4.0 \times 10^{-3}$	0.981
22	$2.5 \times 10^{-4}$	$4.7 \times 10^{-3}$	$3.1 \times 10^{-2}$	$1.9 \times 10^{-5}$	$1.5 \times 10^{-2}$	$4.0 \times 10^{-3}$	0.981
23	$5.0 \times 10^{-4}$	$9.5 \times 10^{-3}$	$3.1 \times 10^{-2}$	$2.2 \times 10^{-5}$	$1.4 \times 10^{-2}$	$4.3 \times 10^{-3}$	0.981

<sup>a</sup>Designation of streams as shown in Fig. A-1 in the Appendix.

Table 5.8. System Iodine DFs for Basic Iodine Behavior Runs<sup>a</sup>

Run	Primary Iodine System DF		Secondary Iodine Removal, M <sub>6</sub> /M <sub>5</sub>	Vessel Off Gas Treatment		Final Off Gas Treatment		Total Plant DF, A7/O1
	Actual, FI/F <sub>1</sub>	Effective, A7/M <sub>6</sub>		Actual M1, N1 + N2 + N3 + OA <sub>1</sub>	Effective, M1/N <sub>1</sub>	Actual, NL/(O1-O1A)	Effective, NL/O1	
1	2.0 x 10 <sup>7</sup>	100	1.1 x 10 <sup>4</sup>	760	1.4 x 10 <sup>3</sup>	1.9 x 10 <sup>4</sup>	330	1.3 x 10 <sup>12</sup>
2	2.0 x 10 <sup>7</sup>	97	1.4 x 10 <sup>4</sup>	960	9.2 x 10 <sup>3</sup>	1.3 x 10 <sup>4</sup>	66	8.2 x 10 <sup>11</sup>
3	2.0 x 10 <sup>7</sup>	195	1.4 x 10 <sup>4</sup>	760	1.9 x 10 <sup>4</sup>	3.7 x 10 <sup>3</sup>	25	1.3 x 10 <sup>12</sup>
4	2.0 x 10 <sup>7</sup>	97	1.4 x 10 <sup>4</sup>	960	1.0 x 10 <sup>3</sup>	7.5 x 10 <sup>3</sup>	530	7.7 x 10 <sup>11</sup>
4A	2.0 x 10 <sup>7</sup>	97	1.4 x 10 <sup>4</sup>	960	1.0 x 10 <sup>3</sup>	380	530	3.3 x 10 <sup>11</sup>
5	2.0 x 10 <sup>7</sup>	92	1.3 x 10 <sup>4</sup>	1.0	9.4 x 10 <sup>3</sup>	1.0 x 10 <sup>3</sup>	61	6.9 x 10 <sup>11</sup>
5A	2.0 x 10 <sup>7</sup>	92	1.3 x 10 <sup>4</sup>	1.0	9.4 x 10 <sup>3</sup>	52	61	3.3 x 10 <sup>11</sup>
6	2.0 x 10 <sup>7</sup>	97	1.3 x 10 <sup>4</sup>	0.94	9.5	2.2 x 10 <sup>4</sup>	1.6 x 10 <sup>4</sup>	2.0 x 10 <sup>11</sup>
6A	2.0 x 10 <sup>7</sup>	97	1.3 x 10 <sup>4</sup>	0.94	9.5	1.1 x 10 <sup>3</sup>	1.6 x 10 <sup>4</sup>	1.3 x 10 <sup>10</sup>
7	2.0 x 10 <sup>7</sup>	84	1.4 x 10 <sup>4</sup>	0.94	0.94	1.2 x 10 <sup>3</sup>	1.2 x 10 <sup>3</sup>	1.3 x 10 <sup>9</sup>
10	1.9 x 10 <sup>7</sup>	33	1.4 x 10 <sup>4</sup>	0.99	0.99	1.2 x 10 <sup>3</sup>	1.2 x 10 <sup>3</sup>	5.4 x 10 <sup>8</sup>
11	2.0 x 10 <sup>7</sup>	103	1.4 x 10 <sup>4</sup>	745	1.8 x 10 <sup>4</sup>	3.7 x 10 <sup>3</sup>	25	6.8 x 10 <sup>11</sup>
14	2.0 x 10 <sup>7</sup>	84	1.4 x 10 <sup>4</sup>	1.2	0.94	1.2 x 10 <sup>3</sup>	1.2 x 10 <sup>3</sup>	1.3 x 10 <sup>9</sup>
15	2.0 x 10 <sup>7</sup>	98	1.4 x 10 <sup>4</sup>	1.8	1.8 x 10 <sup>3</sup>	2.0 x 10 <sup>4</sup>	329	8.1 x 10 <sup>11</sup>
18	2.0 x 10 <sup>7</sup>	59	1.4 x 10 <sup>4</sup>	6.5	1.0	1.2 x 10 <sup>3</sup>	1.2 x 10 <sup>3</sup>	9.7 x 10 <sup>8</sup>
19	1.9 x 10 <sup>7</sup>	30	1.4 x 10 <sup>4</sup>	2.1	2.1 x 10 <sup>3</sup>	2.0 x 10 <sup>4</sup>	329	3.0 x 10 <sup>11</sup>
22	1.9 x 10 <sup>7</sup>	25	1.4 x 10 <sup>4</sup>	1.1	1.1	1.2 x 10 <sup>3</sup>	1.2 x 10 <sup>3</sup>	4.7 x 10 <sup>8</sup>
23	1.9 x 10 <sup>7</sup>	17	1.4 x 10 <sup>4</sup>	1.1	1.1	1.2 x 10 <sup>3</sup>	1.2 x 10 <sup>3</sup>	3.1 x 10 <sup>8</sup>

<sup>a</sup>Streams are designated as shown in Fig. A-1 in the Appendix.

Runs 10 and 11 represent duplication of runs 7 and 3, except that the iodine escaping to the storage cell and mechanical handling cell was increased by factors of 3.3 and 2 respectively. Overall plant retention factors were reduced by factors of 2.4 and 1.2 respectively. The reduction was less for the case of the higher initial DF (run 3), again due to the limiting effect of recontamination.

The main evolution of  $I_2$  takes place in the feed adjustment step. In runs 1-7, it was assumed that 99.9% of the iodine in solution reports to the off-gas stream, which then feeds the primary iodine removal system. Two series of runs were made to investigate the effect of  $I_2$  evolution (99.0% and 95.0%) in the feed adjustment step. The same basic air flow rates that were used in runs 1 and 7 were also used in this series of runs. Any iodine not evolved during the feed adjustment step remained in the feed stream to the solvent extraction system. The major portion of the iodine in the liquid system finds its way to the first-cycle acid-water recovery system, causing a significant increase in the iodine inventory (Table 5.7). The iodine retained by the resins in the acid-water recovery systems is released during burning in the waste handling sections and eventually reports to the off-gas system feeding the secondary iodine removal system. The overall retention factors for iodine are reduced by decreased iodine evolution in the feed adjustment step. When the iodine evolution is reduced from 99.9 to 99.0%, the overall DF will be decreased by a factor of 1.3 to 1.5; for a reduction from 99.0 to 95.0%, the DF will be further decreased by a factor of 2.1 to 2.5.

Reducing the iodine evolution during feed adjustment increases the amount of iodine that bypasses the primary iodine removal system, causing an increase in the load placed on the secondary iodine removal system and resulting in a proportional increase in the amount of iodine released in the stack gas.

One additional run (run 23) was made in an effort to combine the effect of high head-end release with reduced evolution in the feed adjustment step. The air flow rates for this run were the same as those used in run 7, in which a significant amount of the off-gas from the head-end sections bypassed the primary iodine removal system. The results show a

significant decrease in the amount of iodine reporting to the primary iodine removal system and a corresponding increase in the feed to the secondary iodine removal system. The amounts of iodine observed in the other gas streams are as one would expect; the overall DF was reduced by a factor of  $\sim 4$ .

A series of runs was made in which the various iodine removal systems were either deleted from the system or their removal efficiency was reduced. Four basic air flow patterns were used in this part of the study. In each run, the head-end release rate of iodine was held constant at the standard value and an iodine evolution rate of 99.0% was assumed in the feed adjustment step. The secondary Kr-Xe removal equipment was not operated.

Runs were made with each air flow rate pattern, using the following conditions:

- (1) No primary iodine removal system.
- (2) No secondary iodine removal system.
- (3) Iodine removal efficiency of resins and charcoal beds in acid-water recovery systems reduced from 99.0 to 90.0%.
- (4) Iodine removal efficiency of silver-zeolite beds in off-gas systems reduced from 99.9 to 99.0%.

Table 5.9 lists the conditions used for this series of runs. The distribution of  $^{129}\text{I}_2$  in the various off-gas streams is shown in Table 5.10; the DFs for the individual systems are shown in Table 5.11.

When the primary iodine system is not operated, the iodine that would normally be removed reports to the secondary iodine removal system. For example in run 24, the feed stream for the secondary iodine removal system contains 96 times the iodine present in the corresponding stream for run 15. While the secondary system can remove the bulk of the iodine, the amount of iodine in the feed to the vessel off-gas system for run 24 is 96 times that in run 15. The increased concentrations of iodine in the vessel off-gas and final off-gas systems are partly negated by the effect of recontamination by cell air inleakage and recycle water, and the overall

Table 5.9. Run Conditions Testing the Effect of Reduced  $^{129}\text{I}_2$  Removal Capability<sup>a</sup>

Run	Basic Air Flow Pattern Used in Run	Use of Primary $\text{I}_2$ Removal System?	Use of Secondary $\text{I}_2$ Removal System?	Fraction $\text{I}_2$ Retained by Aqueous	Fraction $\text{I}_2$ Retained by Gas-Phase Beds
15	1	Yes	Yes	0.99	0.999
16	4A	Yes	Yes	0.99	0.999
17	5A	Yes	Yes	0.99	0.999
18	7	Yes	Yes	0.99	0.999
24	1	No	Yes	0.99	0.999
25	4A	No	Yes	0.99	0.999
26	5A	No	Yes	0.99	0.999
27	7	No	Yes	0.99	0.999
28	1	Yes	No	0.99	0.999
29	4A	Yes	No	0.99	0.999
30	5A	Yes	No	0.99	0.999
31	7	Yes	No	0.99	0.999
32	1	Yes	Yes	0.90	0.999
33	4A	Yes	Yes	0.90	0.999
34	5A	Yes	Yes	0.90	0.999
35	7	Yes	Yes	0.90	0.999
36	1	Yes	Yes	0.99	0.990
37	4A	Yes	Yes	0.99	0.990
38	5	Yes	Yes	0.99	0.990
39	7	Yes	Yes	0.99	0.990

<sup>a</sup>Standard head-end release rates, 99%  $\text{I}_2$  evolution in feed adjustment step and no secondary Kr-Xe removal applied to all runs.

Table 5.10. Distribution of  $^{129}\text{I}_2$  for Reduced Efficiency for Various Iodine Removal Systems<sup>a</sup>  
(Fraction of Input)

Run	Feed Primary $\text{I}_2$ Removal, F1	Off-Gas Primary $\text{I}_2$ Removal, M6	Feed Secondary $\text{I}_2$ Removal, M6	Off-Gas Secondary $\text{I}_2$ Removal M1	Recycle Air N2+N3	Feed Final Off-Gas Treatment, N1	Off-Gas Final O-G Treatment, O1	Secondary Recycle Acid-H <sub>2</sub> O Inventory, S1+S2
15	0.9735	$4.91 \times 10^{-8}$	$1.02 \times 10^{-2}$	$7.20 \times 10^{-7}$	$3.54 \times 10^{-10}$	$4.06 \times 10^{-10}$	$1.23 \times 10^{-12b}$	$1.47 \times 10^{-6}$
24	0.9736	0.9735	0.9838	$6.89 \times 10^{-5}$	$2.48 \times 10^{-8}$	$2.85 \times 10^{-8}$	$2.60 \times 10^{-11}$	$1.48 \times 10^{-6}$
28	0.9735	$4.87 \times 10^{-8}$	$1.16 \times 10^{-2}$	$1.16 \times 10^{-2}$	$4.18 \times 10^{-6}$	$4.79 \times 10^{-6}$	$4.36 \times 10^{-9}$	$1.92 \times 10^{-6}$
32	0.9736	$4.91 \times 10^{-8}$	$1.01 \times 10^{-2}$	$8.86 \times 10^{-7}$	$9.96 \times 10^{-9}$	$1.14 \times 10^{-8}$	$1.44 \times 10^{-11}$	$5.91 \times 10^{-6}$
36	0.9735	$4.91 \times 10^{-8}$	$1.02 \times 10^{-2}$	$7.20 \times 10^{-7}$	$3.54 \times 10^{-9}$	$4.06 \times 10^{-9}$	$3.37 \times 10^{-11}$	$1.47 \times 10^{-6}$
16	0.9686	$4.91 \times 10^{-8}$	$1.54 \times 10^{-2}$	$1.09 \times 10^{-6}$	$9.27 \times 10^{-11}$	$9.55 \times 10^{-10}$	$3.79 \times 10^{-12b}$	$1.47 \times 10^{-6}$
25	0.9686	0.9686	0.9841	$6.89 \times 10^{-5}$	$4.72 \times 10^{-9}$	$4.86 \times 10^{-8}$	$4.11 \times 10^{-11}$	$1.48 \times 10^{-6}$
29	0.9686	$4.84 \times 10^{-8}$	$1.75 \times 10^{-2}$	$1.75 \times 10^{-2}$	$1.19 \times 10^{-6}$	$1.23 \times 10^{-6}$	$1.01 \times 10^{-9}$	$2.15 \times 10^{-6}$
33	0.9686	$4.91 \times 10^{-8}$	$1.54 \times 10^{-2}$	$1.99 \times 10^{-6}$	$2.00 \times 10^{-9}$	$2.06 \times 10^{-8}$	$1.53 \times 10^{-10}$	$5.92 \times 10^{-6}$
37	0.9686	$4.91 \times 10^{-8}$	$1.54 \times 10^{-2}$	$1.09 \times 10^{-6}$	$9.27 \times 10^{-10}$	$9.55 \times 10^{-9}$	$9.11 \times 10^{-11}$	$1.47 \times 10^{-6}$
17	0.9681	$4.91 \times 10^{-8}$	$1.60 \times 10^{-2}$	$1.20 \times 10^{-6}$	$1.08 \times 10^{-6}$	$1.19 \times 10^{-10}$	$3.79 \times 10^{-12b}$	$1.48 \times 10^{-6}$
26	0.9681	0.9681	0.9842	$6.90 \times 10^{-5}$	$5.04 \times 10^{-5}$	$5.59 \times 10^{-9}$	$3.98 \times 10^{-11}$	$1.48 \times 10^{-6}$
30	0.968	$4.84 \times 10^{-8}$	$1.00 \times 10^{-1}$	$7.31 \times 10^{-2}$	$7.31 \times 10^{-2}$	$8.11 \times 10^{-6}$	$5.61 \times 10^{-8}$	$5.35 \times 10^{-6}$
34	0.9681	$4.91 \times 10^{-8}$	$1.59 \times 10^{-2}$	$9.42 \times 10^{-6}$	$2.84 \times 10^{-6}$	$3.09 \times 10^{-9}$	$1.38 \times 10^{-10}$	$5.99 \times 10^{-6}$
38	0.9681	$4.91 \times 10^{-8}$	$1.60 \times 10^{-2}$	$1.20 \times 10^{-6}$	$1.01 \times 10^{-6}$	$1.19 \times 10^{-9}$	$2.84 \times 10^{-11}$	$1.48 \times 10^{-6}$
18	0.9671	$4.91 \times 10^{-8}$	$1.70 \times 10^{-2}$	$1.24 \times 10^{-6}$	$1.47 \times 10^{-11}$	$1.24 \times 10^{-6}$	$1.03 \times 10^{-9}$	$1.48 \times 10^{-6}$
27	0.9671	0.9671	0.9841	$6.89 \times 10^{-5}$	$6.95 \times 10^{-9}$	$5.61 \times 10^{-5}$	$4.60 \times 10^{-8}$	$1.48 \times 10^{-6}$
31	0.9671	$4.84 \times 10^{-8}$	$2.10 \times 10^{-2}$	$2.10 \times 10^{-2}$	$2.11 \times 10^{-7}$	$1.70 \times 10^{-2}$	$1.39 \times 10^{-5}$	$2.78 \times 10^{-6}$
35	0.9671	$4.91 \times 10^{-8}$	$1.69 \times 10^{-2}$	$6.42 \times 10^{-6}$	$3.00 \times 10^{-10}$	$2.94 \times 10^{-5}$	$2.51 \times 10^{-8}$	$5.98 \times 10^{-6}$
39	0.9671	$4.91 \times 10^{-8}$	$1.70 \times 10^{-2}$	$1.24 \times 10^{-6}$	$1.47 \times 10^{-11}$	$1.24 \times 10^{-6}$	$1.03 \times 10^{-9}$	$1.48 \times 10^{-6}$

<sup>a</sup>Streams are designated as shown in Fig. A-1 in the Appendix.

<sup>b</sup>Calculated values assuming no secondary Kr-Xe removal system.

Table 5.11. Effective Iodine System Decontamination Factors  
Obtained at Reduced Iodine Removal Capability<sup>a</sup>

Run No.	Primary Iodine System, M7/M6	Secondary Iodine System, M6/M1	Vessel Off-Gas Treatment, M1/N1	Final Off-Gas Treatment, N1/O1	Total Plant DF, A7/O1	Reduction in Total DF (Ratio to Base Run)
15	98	$1.4 \times 10^4$	$1.8 \times 10^3$	$3.3 \times 10^2$	$8.1 \times 10^{11}$	1.0
24	1.0	$1.4 \times 10^4$	$2.4 \times 10^3$	$1.1 \times 10^3$	$3.8 \times 10^{10}$	21
28	86	1.0	$2.4 \times 10^3$	$1.1 \times 10^3$	$2.3 \times 10^9$	$3.5 \times 10^3$
32	98	$1.1 \times 10^4$	78	$7.9 \times 10^2$	$6.9 \times 10^{10}$	12
36	98	$1.4 \times 10^4$	$1.8 \times 10^2$	$1.2 \times 10^2$	$3.0 \times 10^{10}$	27
16	65	$1.4 \times 10^4$	$1.1 \times 10^3$	$2.5 \times 10^2$	$2.6 \times 10^{11}$	1.0
25	1.0	$1.4 \times 10^4$	$1.4 \times 10^3$	$1.2 \times 10^3$	$2.4 \times 10^{10}$	11
29	57	1.0	$1.4 \times 10^3$	$1.2 \times 10^3$	$1.0 \times 10^8$	$2.6 \times 10^3$
33	65	$7.7 \times 10^3$	97	$1.3 \times 10^2$	$6.5 \times 10^9$	40
37	65	$1.4 \times 10^4$	$1.1 \times 10^2$	$1.0 \times 10^2$	$1.1 \times 10^{10}$	24
17	62	$1.3 \times 10^4$	$1.0 \times 10^4$	31	$2.6 \times 10^{11}$	1
26	1.0	$1.4 \times 10^4$	$1.2 \times 10^4$	$1.4 \times 10^2$	$2.5 \times 10^{10}$	10
30	10	1.0	$1.2 \times 10^4$	$1.5 \times 10^2$	$1.8 \times 10^7$	$1.4 \times 10^4$
34	62	$1.7 \times 10^3$	$3.0 \times 10^3$	22	$7.2 \times 10^9$	34
38	62	$1.3 \times 10^4$	$1.0 \times 10^3$	42	$3.5 \times 10^{10}$	7
18	59	$1.4 \times 10^4$	1.0	$1.2 \times 10^3$	$9.7 \times 10^8$	1
27	1.0	$1.4 \times 10^4$	1.2	$1.2 \times 10^3$	$2.2 \times 10^7$	44
31	48	1.0	1.2	$1.2 \times 10^3$	$7.2 \times 10^4$	$1.3 \times 10^4$
35	59	$2.6 \times 10^3$	0.2	$1.2 \times 10^3$	$4.0 \times 10^7$	24
39	59	$1.4 \times 10^4$	1.0	$1.2 \times 10^2$	$9.7 \times 10^7$	10

<sup>a</sup>Streams are designated as shown in Fig. A-1 in Appendix.

plant retention factor for run 24 is only a factor of 21 less than that for run 15. Similar effects are observed in the following run pairs: 16-25, 17-26, and 18-27.

The deletion of the secondary iodine removal system has a very detrimental effect on the plant retention factor for iodine. The removal of the secondary system causes an increase in the feed to the vessel off-gas system by a factor of  $1.6 \times 10^4$  to  $8.33 \times 10^4$ . This is a much larger increase than that observed for the cases in which the primary iodine system was removed ( $I_2$  increased by a factor of 57 to 96). Even with small gains in the DFs of the vessel off-gas system and the final off-gas system, the overall plant retention factors were reduced by factors of  $2.6 \times 10^3$  to  $1.4 \times 10^4$  for runs 28-31.

In the acid-water recovery systems, the aqueous streams are treated for iodine removal by resin and charcoal beds. In runs 32-35, it was assumed that the fraction of  $I_2$  retained by the individual beds was reduced from 0.99 to 0.90. This caused an increase (by a factor of 40) in the amount of  $I_2$  in the recycle acid and water streams, which, in turn, increased the amount of recontamination that took place in the secondary  $I_2$  removal, vessel off-gas, and final off-gas treatment systems. Increased recontamination resulted in a significant decrease in the retention factors for these systems. The overall plant retention factors were reduced by factors of 12 to 40.

Reducing the fraction of iodine retained by the silver zeolite beds in the vessel off-gas system and the final off-gas system from 0.999 to 0.99 has only a small effect on the iodine released in the stack gas. In run 39, the net off-gas is not treated for iodine removal in the vessel off-gas; hence the plant retention factor for iodine was decreased by only a factor of 10 in this run. The retention factors of the vessel off-gas and the final off-gas treatment systems were each reduced by a factor of 10 in runs 36-38. However, the overall plant retention was reduced by only a factor of 7 to 27. These smaller-than-expected reductions in the DF are caused by the relative importance of recontamination by the recycle water stream. For example, in run 36, where the amount of iodine in the

recycle water stream is small compared to that in the gas stream, the apparent DF was reduced by a factor of 27. On the other hand, in run 38, where the amount of iodine in the recycle water stream exceeds that in the gas stream, the apparent DF was reduced by only a factor of 7.

#### 5.4 Distribution of Other Fission Products

An attempt was made to model the behavior of particulate and semi-volatile fission products throughout the process. The modeling in this area is probably the least accurate of any of the components considered; thus the presentation of detailed results is not warranted. In general, the particulate activity will distribute throughout the process off-gas systems similar to the distribution experienced in current-generation plants. Concentrations in the off-gas from the vessel off-gas treatment system will be similar to concentrations in present-generation stack effluents.

Because the gas flow rates are small, use of high-pressure drop filter media purported to have improved particle removal capability might be practical and result in a reduction in particulate concentration in the plant off-gas. Low net gas effluent rates, possibly in combination with improved filtration may result in reducing particulate and semi-volatile activity releases by several orders of magnitude below that obtained in current practice.

#### 6. REFERENCES

1. C. D. Watson et al., A Study of the Reprocessing of Spent Fast Test Reactor Fuel (and Other LMFBR Fuels) in the Nuclear Fuel Services Plant, ORNL-TM-2906 (August 1971) Section 9.
2. Staff of the Oak Ridge National Laboratory, Siting of Fuel Reprocessing Plants and Waste Management Facilities, ORNL-4451 (July 1970) Section 4.1.3.
3. J. P. Nichols and F. T. Binford, Status of Noble Gas Removal and Disposal, ORNL-TM-3515 (August 1971).



## 7. APPENDIX

The general flowsheet used in developing the mathematical model is illustrated in Fig. A-1. Each of 23 process and effluent steps and 6 cell containment enclosures was modeled with respect to incoming streams, internal interactions, and exit streams. Reference drawing numbers for each of the 29 systems modeled are listed in Table A-1, and representative drawings are included for the tritium removal system (Fig. A-3), iodine evolution and feed adjustment (Fig. A-5), and primary iodine removal (Fig. A-7). Typical process flow diagrams were developed for most of the systems modeled, and examples are included for the tritium removal system (Fig. A-2), iodine evolution system (Fig. A-4), primary iodine removal (Fig. A-6), secondary iodine removal (Fig. A-8), and the acid and water recovery systems (Fig. A-9).

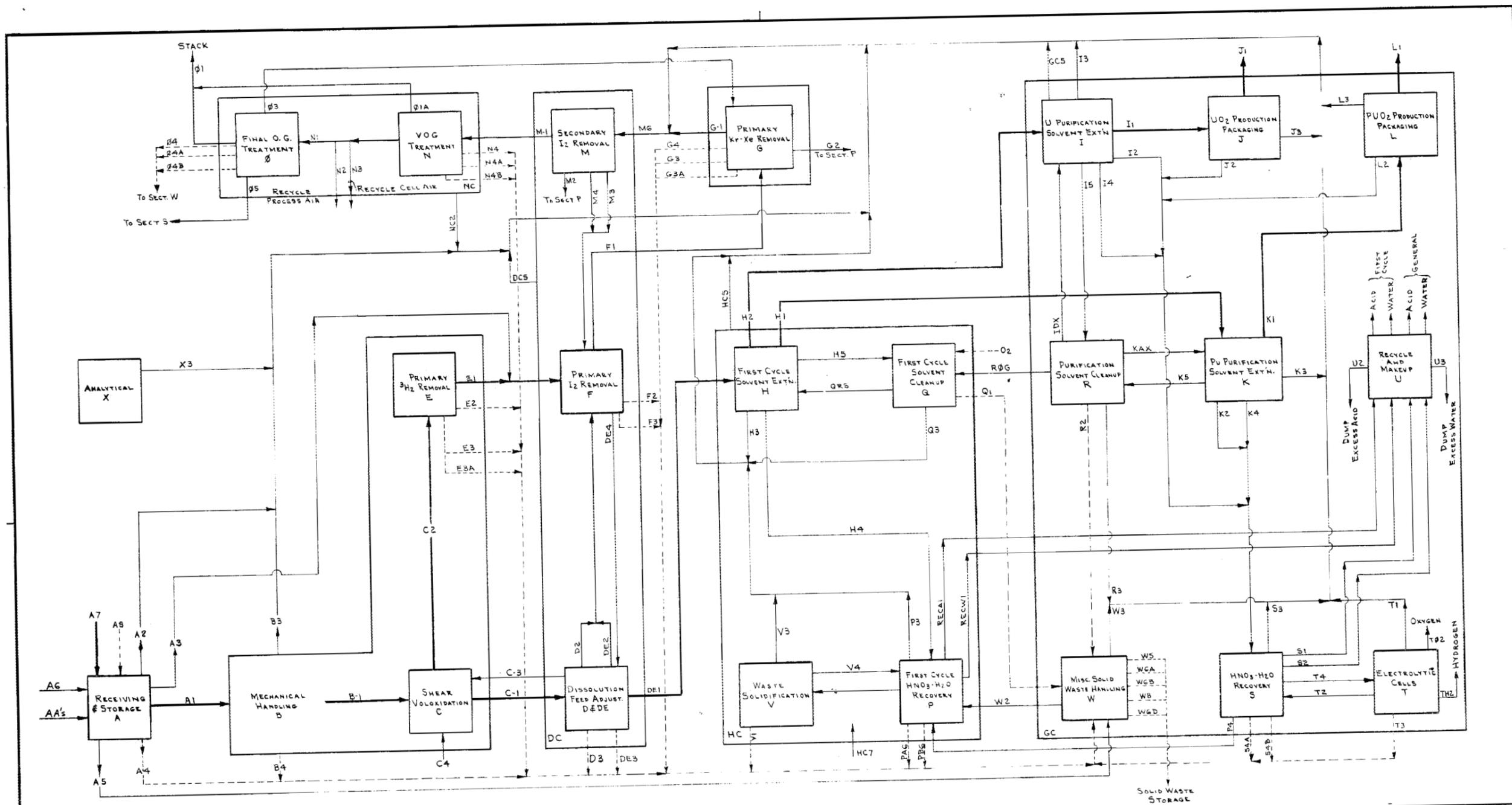
Table A-1. Reference Drawings of Mathematical Models  
of Individual Systems

Dwg. No.	Title
M-12251-CD-022	Mathematical Model Sect. A: Receiving and Storage
M-12251-CD-023	Mathematical Model Sect. B: Mathematical Handling Cell
M-12251-CD-038	Mathematical Model Sect. C: Shear Voloxidizer
M-12251-CD-019	Mathematical Model Sect. D: Dissolver
M-12251-CD-043	Mathematical Model Sect. DC: Dissolver and Iodine Equipment Cell
M-12251-CD-026	Mathematical Model Sect. DE: Iodine Evolution and Feed Adjustment
M-12251-CD-025	Mathematical Model Sect. E: Primary Tritium Removal
M-12251-CD-042	Mathematical Model Sect. F: Primary Iodine Removal
M-12251-CD-039	Mathematical Model Sect. G: Primary Kr-Xe Removal
M-12251-CD-027	Mathematical Model Sect. GC: General Cell
M-12251-CD-035	Mathematical Model Sect. H: First Cycle Solvent Extraction
M-12251-CD-037	Mathematical Model Sect. HC: Solvent Extraction Cell 1st Cycle
M-12251-CD-040	Mathematical Model Sect. I: U Purification SX
M-12251-CD-028	Mathematical Model Sect. I: UO <sub>2</sub> Production and Packaging
M-12251-CD-041	Mathematical Model Sect. K: Pu Purification SX
M-12251-CD-034	Mathematical Model Sect. L: PuO <sub>2</sub> Production and Packaging
M-12251-CD-030	Mathematical Model Sect. M: Secondary Iodine Removal System
M-12251-CD-046	Mathematical Model Sect. N: Vessel Off-Gas
M-12251-CD-053	Mathematical Model Sect. NC: Final Off-Gas Treatment Cell
M-12251-CD-048	Mathematical Model Sect. Ø: Final Off-Gas Treatment

Table A-1 - (Continued )

M-12251-CD-031	Mathematical Model Sect. P:	First Cycle Acid and Water Recovery
M-12251-CD-029	Mathematical Model Sect. Q:	1st Cycle Solvent Cleanup
M-12251-CD-032	Mathematical Model Sect. R:	Purification Cycles Solvent Cleanup
M-12251-CD-036	Mathematical Model Sect. S:	General Acid-Water Recovery
M-12251-CD-051	Mathematical Model Sect. T:	Hydrogen-Oxygen Production
M-12251-CD-045	Mathematical Model Sect. U:	Makeup Water and Acid
M-12251-CD-033	Mathematical Model Sect. V:	Waste Solidification
M-12251-CD-047	Mathematical Model Sect. W:	Solid Waste Handling
M-12251-CD-052	Mathematical Model Sect. X:	Analytical

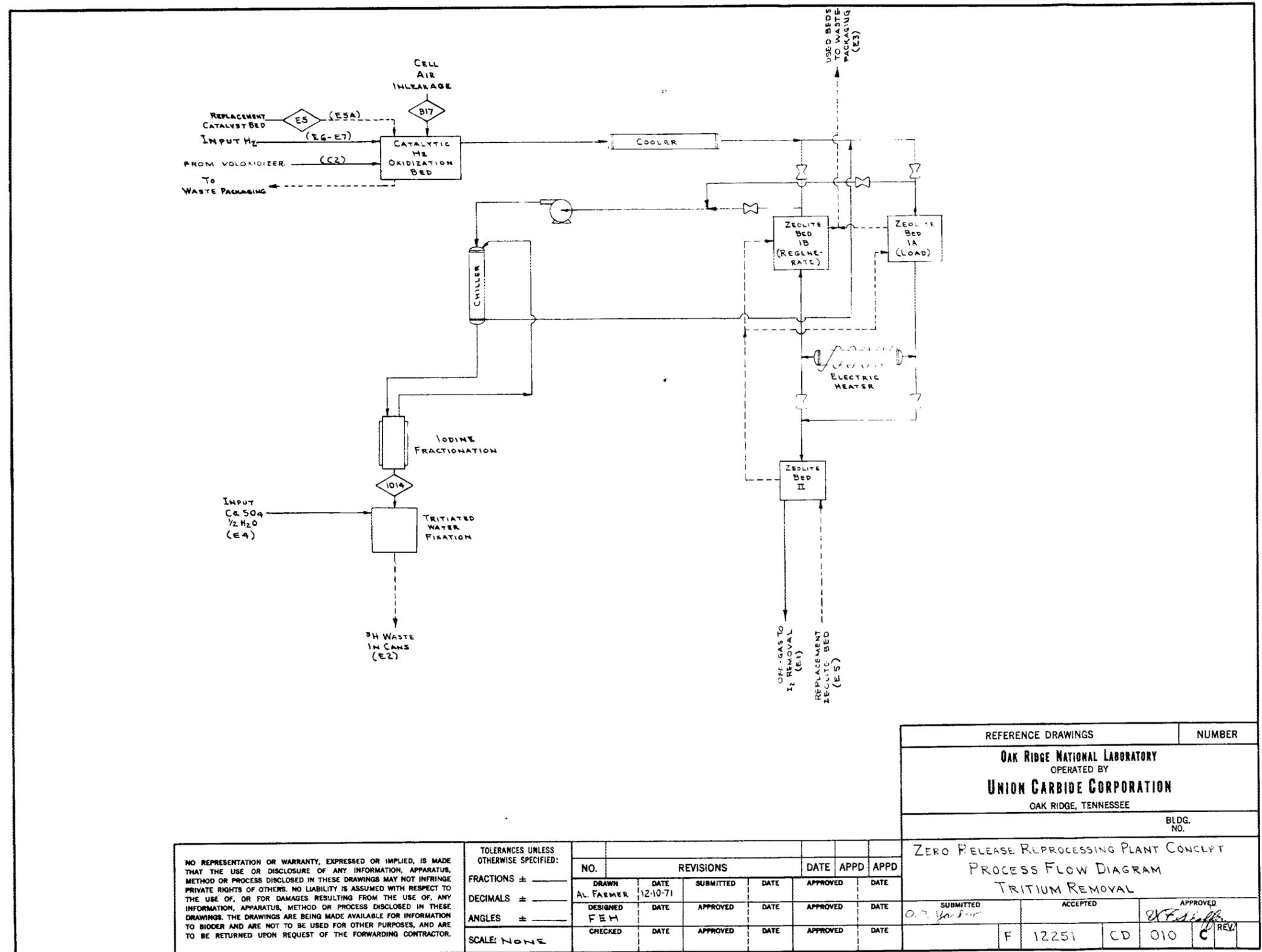
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NOTES:  
 (1) SEE INDIVIDUAL MATHEMATICAL MODELS FOR MORE DETAILS, I.E. -022-C FOR SECT A, -045-C FOR SECT U.  
 (2) MAIN PROCESS FLOW INDICATED HEAVY LINES, I.E. A7, A1, B1, C1 --- J1, L1. DOTTED LINES INDICATE FLOW OF SOLIDS.  
 (3) SIMILAR DWG. FOR AIR FLOW IS

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SCALE:											

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SIMPLIFIED BLOCK FLOW DIAGRAM ZERO RELEASE REPROCESSING PLANT		
SUBMITTED	ACCEPTED	APPROVED
O.O. Yarbrough		
F 12251	CD 018	D

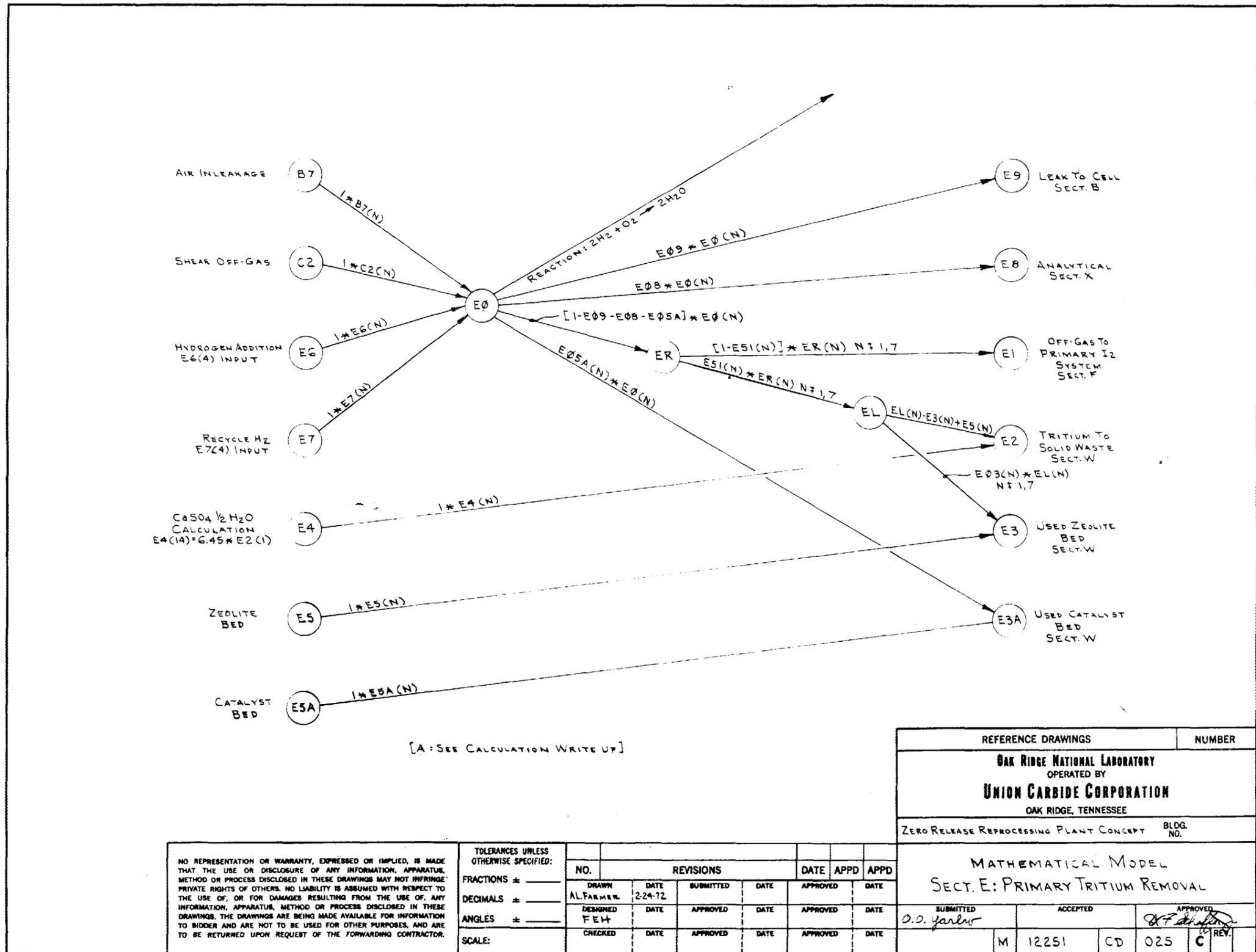


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REV. <i>C</i>	

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	FEH				
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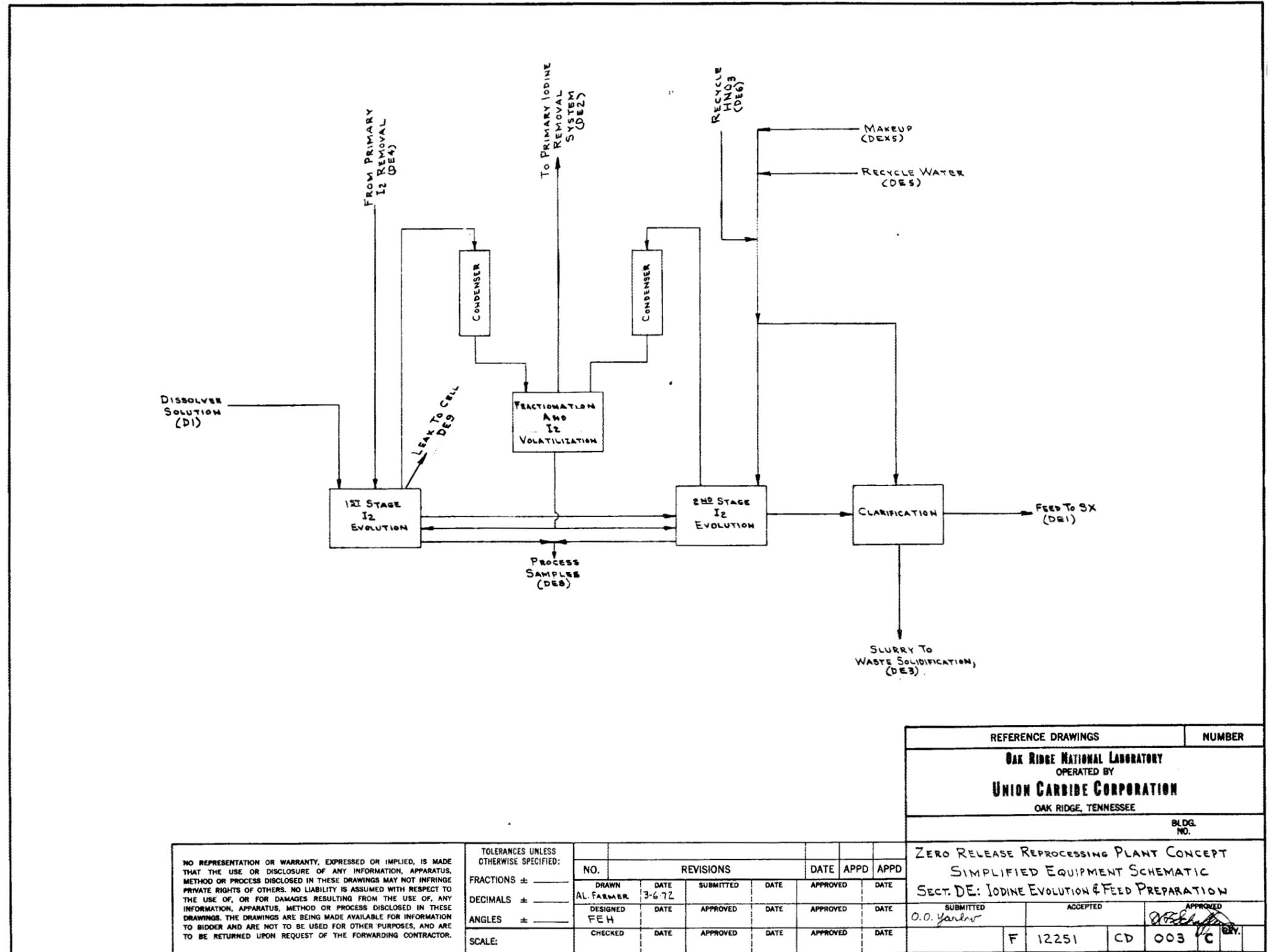


[A: SEE CALCULATION WRITE UP]

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<b>MATHEMATICAL MODEL</b> <b>SECT. E: PRIMARY TRITIUM REMOVAL</b>	
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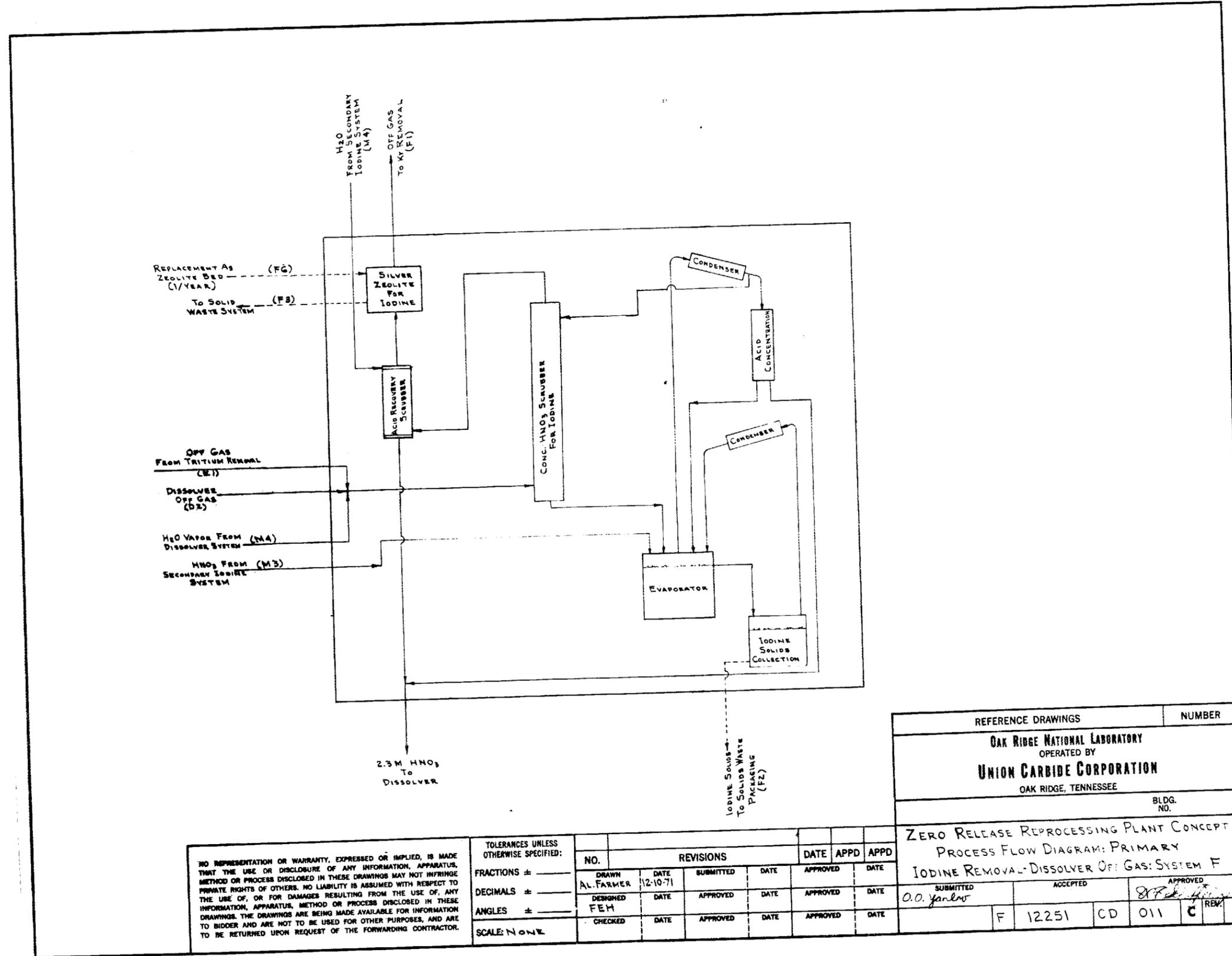
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ZERO RELEASE REPROCESSING PLANT CONCEPT					
SIMPLIFIED EQUIPMENT SCHEMATIC					
SECT. DE: IODINE EVOLUTION & FEED PREPARATION					
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O.O. Yarnor				<i>[Signature]</i>	
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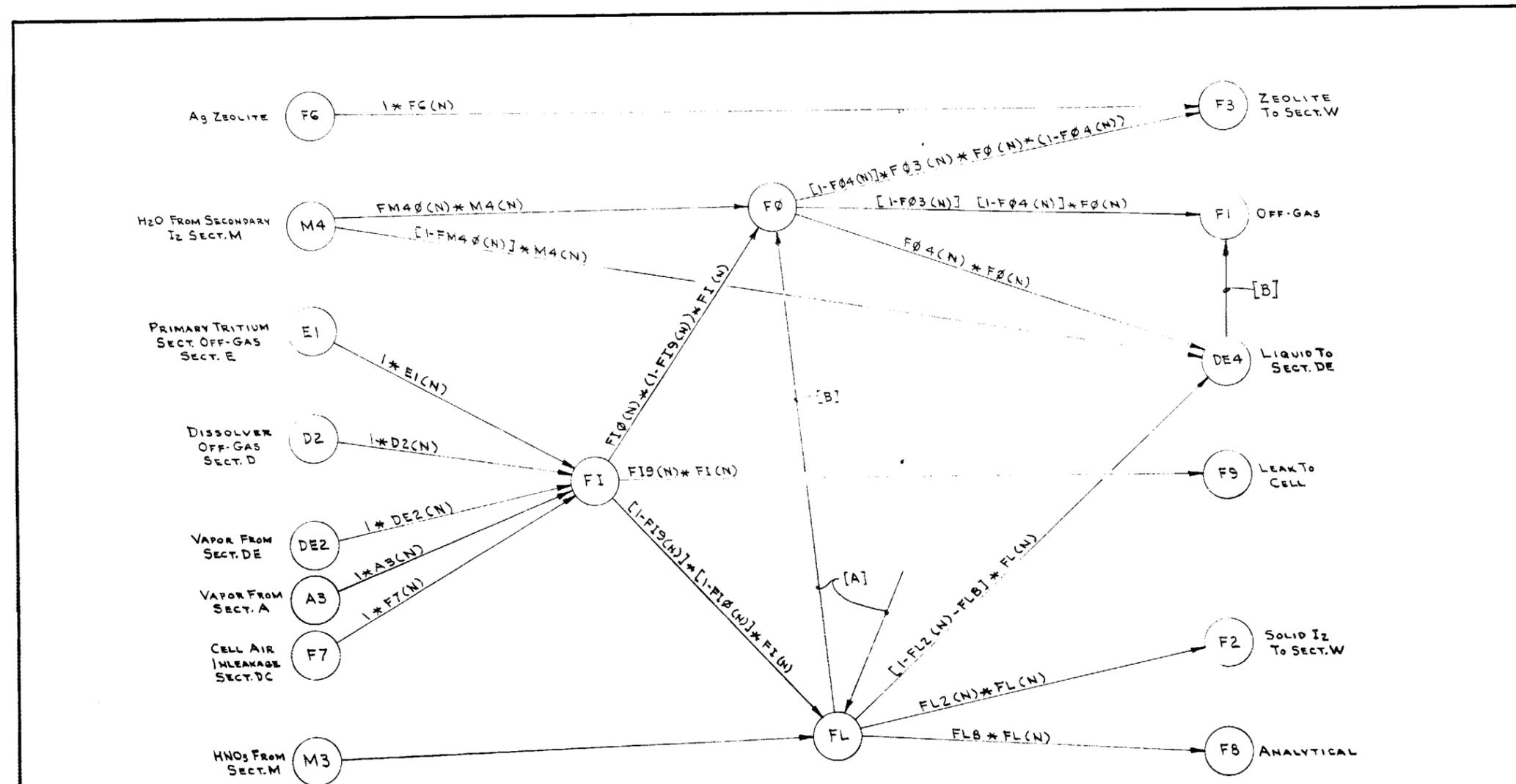




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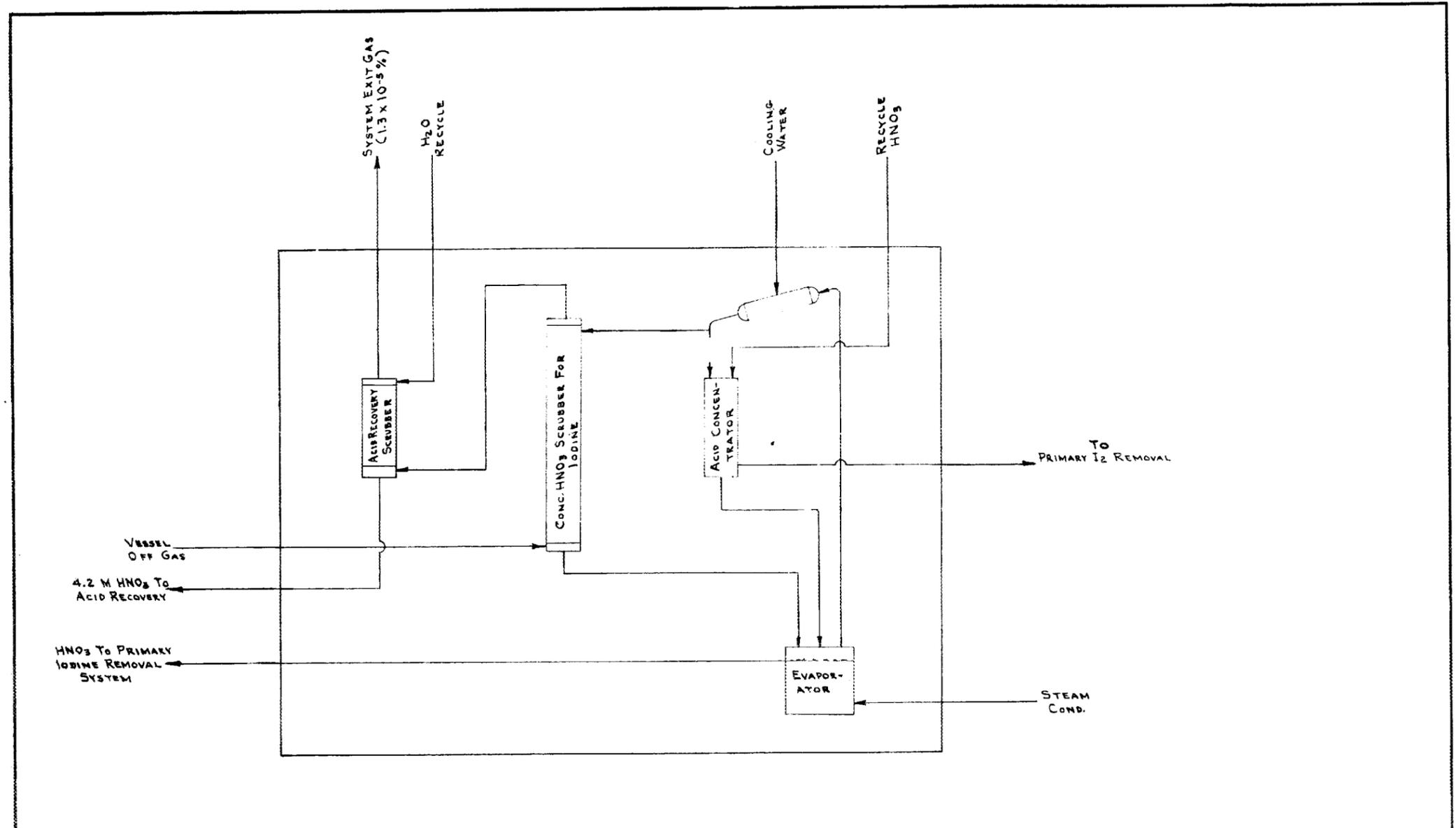
[A] SEE APPENDIX FOR REACTIONS FOR IODINE  
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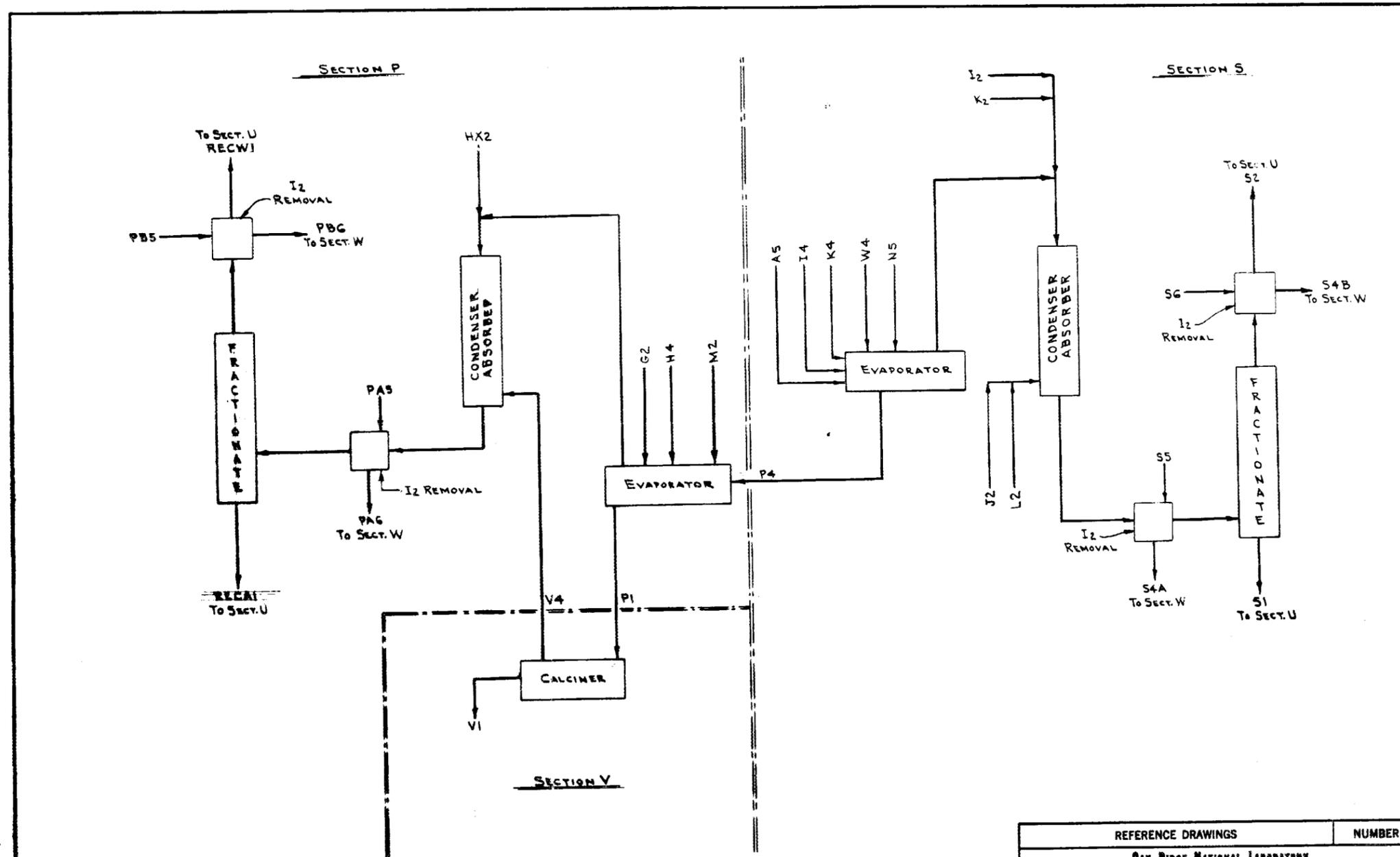


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NO.	REVISIONS				DATE	APPD	APPD
	DRAWN	DATE	SUBMITTED	DATE			
1	AL. FARMER	3-13-72					
	DESIGNED	DATE	APPROVED	DATE	APPROVED	DATE	
	FEH						
	CHECKED	DATE	APPROVED	DATE	APPROVED	DATE	

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