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SUBJECT: Survey of Processes for Control of Xe-135 Poisoning in HRE-3

TO: W. R. Gall

FROM: E. A. Mason

SUMMARY

Various systems for reducing the effect of poisoning by Xe-135 in HRE-3 have been evaluated. Considering the design criteria that the reactor shall not be dependent on operation of auxiliary equipment, xenon removal by means of direct let-down of either vapor or fuel solution was eliminated. Processing of condensate produced in the high pressure system for removal of iodine appears presently to be the most satisfactory method (See Fig. 9). Processing of 1.5 gallons per minute of condensate should result in reduction of xenon poison fraction to about 1%. The proposed system combines condensate production and prevention of xenon poisoning so that the need for let-down and pump-up between high and low pressures is reduced and the overall reactor design is thereby simplified.

Development work is required before a final safe design can be specified. Information is needed concerning: the explosive limits of mixtures of deuterium, oxygen and heavy water vapor; the recombination rate of deuterium and oxygen in fuel solution containing copper; the pumping and gas-liquid contacting efficiency of jet eductors operating at reactor conditions; the stability of silver-silver iodide adsorption beds under irradiation and at the temperatures which will be reached in the presence of the decay of radioactive iodine to xenon.

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2.0 Introduction

The design criteria for HRE-3 which have been established state that the reactor shall be a 60 Mw, 2 zone, aqueous homogeneous reactor. The core is to be a solution of uranyl sulfate and heavy water, and the blanket is to be a slurry of thorium oxide and heavy water. Approximately 50 Mw of heat will be generated in the core and 10 Mw in the blanket system. The design temperature at the core outlet has been set at 280°C with a maximum of 300°C. The temperature rise across the reactor has been established at 30°C. Copper catalyst is to be used for internal recombination of radiolytic deuterium and oxygen.

One of the major objectives of HRE-3 is to demonstrate breeding. Another important criteria is that the operation of the reactor shall not depend upon the continuous operation of any auxiliary piece of equipment. This latter criteria is directed particularly towards avoidance of the necessity that feed pumps operate continuously, pumping solution from low pressure to high pressure. The design criteria for avoidance of dependence on the auxiliary system implies that the production of condensate (for purging pumps and for concentration control) will be carried out at reactor temperature and pressure so that it will not be necessary to pump up condensate from a low pressure to a high pressure as is done in the HRT. The operating pressure of the HRE-3 has been established at 1500 psi, and since the maximum solution temperature has been set at 300°C to avoid phase separation of the fuel solution, it is not possible to produce condensate by boiling the fuel solution at the reactor pressure.

Therefore, to produce condensate at reactor pressure it will be necessary to use an inert gas to strip D_2O from the fuel solution and carry it into a condenser where the D_2O is removed from the gas, which would then be recycled to pick up more D_2O . Oxygen appears to be the best choice for such an inert gas due to its compatibility with the fuel solution and the corrosion protection which it affords. Bolger and Maak⁽¹⁾ have discussed a preliminary design for such a system. Since the oxygen will, in effect, act as an oxygen pressurizing system, the need for a steam pressurization system is eliminated (elimination of steam pressurization also decreases somewhat the requirement for condensate production. Use of an oxygen pressurization and condensate production system can lead to explosive conditions in the gas phase due to the stripping of deuterium from the fuel solution. Bolger and Maak⁽¹⁾ and Arthur D. Little⁽²⁾ discuss the problem of deuterium-oxygen explosive conditions in an oxygen pressurization system.

Preliminary nuclear calculations on various HRE-3 systems have been carried out. At the present time HRE-3 reactor has been established to have a 4 ft diameter core with a 9 ft diameter pressure vessel (forming a 2.5 ft thick blanket). Until stress calculations are completed, the core tank thickness has been assumed to be 1/2 in. zircaloy. For a reactor containing 1000 gm/liter ThO_2 in the blanket and a total poison fraction of 7% including 1% xenon (this implies rapid processing to reduce the xenon poison level), the following breeding gains have been calculated: two-group calculations for equilibrium conditions give a breeding gain of 0.11;⁽³⁾

two-group time-dependent calculations indicate an initial breeding gain of approximately 0.16;⁽⁴⁾ and multi-group equilibrium calculations indicate a breeding gain of 0.06.⁽⁴⁾ The xenon poison fraction in the absence of rapid processing for xenon removal has been calculated to be about 5.3%. If HRE-3 were to be run with no processing for xenon removal, the above breeding gains would therefore be reduced by approximately 0.032 to breeding gains of about 0.08, 0.13, and 0.03 for the three calculations listed above in the order given.

These nuclear calculations indicate that processing for xenon removal does not appear to be absolutely necessary in order to demonstrate breeding in HRE-3, although xenon poisoning accounts for a significant portion of the saturated breeding gain. Reduction of the xenon poison level to approximately 1% xenon would add appreciably to the likelihood of success in demonstrating breeding in this reactor when built and operated.

This report contains the results of a series of preliminary evaluations made on methods for reducing the xenon poison level in each HRE-3. Methods surveyed depend upon removal of xenon-135 or its precursor iodine-135. One phase of the work has been directed towards examining the relationships between the reactor system and the xenon processing system to determine the effect each has on the other.

2.1 Xenon Removal by Liquid Letdown

Xenon poisoning in a homogeneous reactor can be reduced by removing fuel solution from the high pressure system, flashing or stripping this fuel solution to remove iodine and xenon, conversion of the stripped iodine to xenon by absorption and decay upon a silver bed, followed by condensation of the vapors so that the liquid may be returned to the high pressure system. Due to the low solubility of xenon in the condensate, little poison would be returned to the reactor in the condensate. The off-gas from the condenser is passed into a disposal system for radiolytic fission-product gas. Such a system is now contemplated for use in the HRT. Figure 1 shows the effect of liquid letdown on the xenon poison fraction in HRE-3. The minimum xenon poison fraction obtainable with liquid letdown is shown, since the condensate returned to the high pressure system was assumed to contain no xenon or iodine (whereas actually a small amount of xenon and iodine would be contained). Slightly more than 7 gal/min. of fuel solution would be let down in order to achieve a xenon poison fraction in the reactor of 0.01.

Use of such a liquid letdown system in HRE-3 is incompatible with the design criteria that dependence on auxiliary components be avoided.

Figure 2 shows that xenon decay in the high pressure gas space of HRE-3 will have a negligible effect upon the xenon poison fraction for any reasonable volume of gas.

2.2 Stripping of Iodine-135 and Xenon-135 from Fuel Solution

The solubility of iodine and xenon in fuel solution at reactor temperature and pressure favors the stripping of the iodine and xenon from the fuel solution into any gas phase present.⁽⁵⁾⁽⁶⁾ However, deuterium and oxygen,

either formed by the radiolytic decomposition of the heavy water present in the fuel solution or by the addition of oxygen to the system for corrosion control or oxygen pressurization, are also relatively insoluble in the fuel solution and tend to strip from the fuel solution into any gas phase present. Thus care must be taken in designing and operating iodine and xenon stripping systems so as to avoid the possibility of producing an explosive mixture of deuterium and oxygen in any gas volume of appreciable size.

The methods which have been considered for the stripping of iodine and xenon from the fuel solution at high pressure can be divided into three categories.

1. Direct letdown of vapor to a low pressure system.
2. Conversion of iodine in the high pressure gas to xenon and letdown of resulting gas.
3. Treatment of condensate produced at high pressure.
 - a. Removal of xenon and iodine from condensate at low pressure.
 - b. Removal of xenon and iodine from condensate in high pressure system.
4. Use of auxiliary heavy water stream to absorb iodine stripped from fuel.

The methods listed above involve either the stripping of iodine and xenon from the fuel solution into the gas phase by the use of bubbles of gas circulating in the core (these gas bubbles may be either deuterium and oxygen formed by radiolytic decomposition or an inert gas such as oxygen pumped into the system) or by the contact of the fuel solution with a stripping gas in a side stream off the main circulating loop, or a combination of both gas bubbles and stripping of fuel solution in a side loop.

The evaluations carried out in the study have been concerned principally with the methods of removing the xenon and iodine from the primary high pressure loop and have not considered the problem of holdup and ultimate disposal of the fission product gases. Equations relating the xenon poison level and the fuel solution to various conditions of reactor operation and flow rates of various process streams have been derived for a number of different methods of stripping the fission product gases from the fuel solution. These equations have been numerically evaluated with a few process rates of practical interest in the HRE-3. The equations will be useful in evaluating the capabilities for xenon removal with any changes in the HRE-3 concept as the design progresses. A few systems have been considered in detail to show the relationship between xenon and iodine stripping, deuterium and oxygen stripping and explosive conditions, concentration of recombination catalyst required, temperature level of reactor, and process flow rates.

2.3 Explosion Limits for Mixtures of Hydrogen-Oxygen-Water Vapor

The design and operation of systems for oxygen pressurization, high pressure condensate production, and xenon and iodine stripping are dependent upon the explosive limits for mixtures of deuterium oxygen and water vapor. Data on deuterium and oxygen are not available and these limits have not even been well established for mixtures containing hydrogen. Data reported by various investigators for the explosive limits of mixtures of hydrogen, oxygen and water vapor vary widely.

In one report⁽⁸⁾, Battelle Memorial Institute stated that in a 3-inch autoclave ignition occurred at 250°C in mixtures of water vapor, hydrogen and oxygen containing as low as about 0.4% stoichiometric mixture of hydrogen and oxygen (Battelle was unable to establish the lower limit in these experiments). In their work on the gas pressurization for the HRT, Arthur D. Little⁽²⁾ reviewed reported data on the explosive limits for hydrogen and oxygen; Pigford⁽¹⁵⁾ also reviewed the literature on hydrogen-oxygen explosive limits. With the exception of the Battelle data quoted above, other workers have found that the explosive limits for hydrogen in air or oxygen are about 4 to 5% hydrogen or greater.

Consideration of the chain stopping effect of the water vapor molecules would lead one to suppose that explosive limits for hydrogen and oxygen saturated with water vapor would be higher than for the dry gases. Earlier work by Battelle⁽⁷⁾ in autoclaves of diameter less than 3 inches confirmed this deduction. The second Battelle report⁽⁸⁾ suggests that the very low values found by them may be due to the greater volume-to-surface ratio employed in the experiments with the 3-inch autoclave; a decrease in the volume-to-surface ratio would be expected to decrease the explosive limits. However, in experiments carried out at atmospheric pressure in two ft diameter ducts on the explosive limits of hydrogen, oxygen, and water vapor, NACA reported⁽⁹⁾ that gases containing less than 9.5% hydrogen were out of the combustible range (the low values reported by Battelle were obtained in a 3-in. diameter autoclave). Judging from the data obtained by Bowen and Townsend⁽¹⁰⁾ studying the effect of explosive limits of hydrogen and oxygen as a function of pressure, the difference between the NACA data and the Battelle work in the 3-in. autoclave cannot be explained on the basis of difference in pressure. Furthermore I. Spiewak⁽¹¹⁾, who was following the work at Battelle, on examination of the raw data reveals there was a strong likelihood that poor mixing of gases was present in the Battelle experiments (3-in. autoclave) so that the actual composition of the gas in the ignition zone was not known. In work by Battelle with a 2-1/2 in. diameter autoclave⁽⁸⁾, the lower explosive limit at 250°C was found when the partial pressure of the stoichiometric hydrogen-oxygen mixtures was about 16% of the total pressure of the gas-water vapor mixture, or when the hydrogen partial pressure was approximately 11% of the total pressure. This value agrees with the limits reported by others for dry air or oxygen and hydrogen plus allowance for an increase in the limits due to the chain-stopping action of the water molecules present in the gas-vapor mixture. Further work on determining the explosive limits of hydrogen

and oxygen in the presence of water vapor is underway at Syracuse University under contract to the Reactor Experimental Engineering Division of ORNL.

At this time selection of a safe lower explosive limit is open to considerable question. If the Battelle data for the 3 in. autoclave are ignored on the basis that they are subject to question in experimental technique, one might use the 10 or 11% hydrogen value obtained in the early work of Battelle or perhaps more safely the 4% hydrogen value reported for dry mixtures of hydrogen and oxygen using the quenching effect of the water molecules as a factor of safety. In any event the reactor system must be designed and operated so as to avoid stagnant gas spaces where concentration of deuterium might be effected by condensation or diffusion effects.

In this work on oxygen pressurization⁽²⁾, Arthur D. Little also concluded that an exact value for the explosion limit cannot be given at this time. In their work, a value of 1% deuterium was assumed apparently as a compromise between the very low hydrogen value of Battelle⁽⁸⁾ (less than 0.4%) and the hydrogen values for dry gases. Since the rate of recombination of hydrogen (and presumably deuterium) and oxygen in fuel solution at a given temperature has been reported to be proportional to the product of the copper and hydrogen concentration, 100% recombination in fuel solution at high specific powers (therefore high decomposition rate per volume solution) can only be achieved by having appreciable quantities of hydrogen in solution (the copper concentration cannot be increased beyond about 0.03M copper without phase separation in the fuel solution). For HRE-3 operating at 1500 psi, 50 Mw in the core, and at a core outlet temperature of 280°C, the partial pressure of deuterium in equilibrium with fuel at the core outlet required to achieve 100% recombination in the main fuel solution is calculated to be approximately 190 psi. Thus vapor in equilibrium with such fuel solution would be unsafe. If the partial pressure of deuterium is decreased below this value of 190 psi by addition of a diluent gas (such as oxygen), then 100% recombination of the decomposition gas will not be achieved on the main fuel circulation loop for the reactor operating conditions listed. Provision for increased hold-up outside the reactor core would enable 100% liquid-phase recombination at lower deuterium partial pressures, but would decrease the specific power.

Data available today on explosion or ignition limits are for mixtures containing hydrogen. Information on the limits for mixtures containing deuterium is not available. The usual (and simplest) assumption is that the limits are the same for hydrogen and deuterium. Considering the importance of preventing explosions in aqueous reactor systems, it is recommended that the explosion limits for mixtures of deuterium, oxygen and water vapor be determined.

3.0. Results

3.1 Letdown of Vapor to a Low Pressure System

Xenon poison level in HRE-3 could be reduced by the letdown of vapor from the high pressure system to a low pressure system. The deuterium and oxygen letdown would be recombined and the resulting gases condensed to recover the D₂O. Presumably the letdown gases would be passed over a silver bed to remove

the iodine prior to entering a recombiner. Thus the iodine would be held up for decay to xenon on the silver bed and very little xenon would dissolve in the condensate returned to the reactor system. Xenon would thus be removed from the system by venting of the condenser off-gases to a radioactive gas disposal system. This method of xenon removal is similar in many respects to that of the HRT. Figure 3-A shows a simplified block-and-line diagram of the high pressure system of such a reactor. (Figure 3 contains a number of simplified black line flow sheets which were used in calculations of the processing rates required for various xenon poison levels in the reactor.)

Figure 4 shows the effect of vapor letdown on the xenon poison level. The xenon poison level for System 3A has been plotted against the amount of D_2O contained in the vapor letdown since this is an indication of the amount D_2O which would have to be pumped up into the high pressure system. In addition to the D_2O vapor which would be condensed there is also the D_2O formed by recombining the D_2 and O_2 in the system. Also shown on Figure 4 is the amount of non-condensable gases (principally deuterium and oxygen) which would be letdown in the vapor. The ratio of deuterium to oxygen in the vapor letdown would be dependent upon the copper concentration in the fuel solution and the oxygen overpressure in the reactor system.

As pointed out by Haubenreich⁽¹²⁾ and Gift⁽¹³⁾ letdown of gas in aqueous homogeneous reactor systems can result in a deficiency of oxygen following the gas separator unless a considerable excess of oxygen is present in the gas entering the separator. Therefore, in addition to the rather large amount of condensate which would have to be pumped back into the reactor system, considerable oxygen would have to be added to the reactor system and then disposed of in an off gas system. In order to prevent oxidation of the charcoal beds, it might be necessary to burn the oxygen with hydrogen prior to admission of the condenser off gas to the charcoal beds (this will depend upon the amount of oxygen being letdown under the system finally selected).

Due to the fact that in a system such as this the vapor and fuel solution are essentially at equilibrium in the reactor system, considerable deuterium will be stripped from the fuel solution. Thus it is necessary to design and operate the system to avoid explosive mixtures. This could be accomplished by the addition of diluent vapor in the low pressure system, as in the HRT, and by avoiding any significant volume of vapor in the high pressure system.

The characteristics of System 3A with a requirement for a significant amount of pump up of condensate do not appear consistent with the design requirement that the reactor operation shall be independent of auxiliary systems. On this basis, System 3A is not recommended for use in HRE-3.

To illustrate the effect of letdown of condensate containing iodine (See Figure 3-B) as opposed to vapor letdown (Figure 3-A), the xenon poison fraction in the HRE-3 is shown in Figure 4 as a function of the condensate letdown from a system in which condensate is produced by cooling in the

high pressure gas phase. In both cases, the general assumption is made that negligible iodine and xenon are returned to the high pressure fuel system. Comparison of the curves for Systems 3A and 3B on Figure 4 show that where the D_2O letdown is small, letdown of condensate produces a lower poison fraction than letdown of vapor. This is due to the higher solubility of iodine in condensate than in fuel solution. However, as the amount of material letdown is increased, the condensate letdown system suffers by the fact that it is very inefficient in removal of xenon, due to the low solubility of xenon in the condensate. The vapor system at all times will remove both iodine and xenon from the high pressure system, whereas the condensate is limited principally to the removal of iodine.

System 3B as shown in Fig. 3 would not be operable for HRE-3 inasmuch as an explosive mixture would exist in the gas space even at the maximum copper concentration which could be tolerated without phase separation when the reactor outlet temperature is $280^{\circ}C$.

3.2 Conversion of Iodine to Xenon in High Pressure System and Removal of Xenon by Letdown of Vapor

Another possible method for controlling the xenon poisoning in HRE-3 consists of stripping iodine and xenon from the fuel solution at high pressure into a gas stream, conversion of the iodine contained in the gas stream to xenon by adsorption and decay on a silver coated bed, followed by letdown of vapor for removal of xenon. Three systems employing this process are shown in Fig. 3, as Systems C, D and E. All three of these systems would produce condensate at high pressure; in system C it is assumed that the fuel solution and gas phase are in equilibrium with respect to dissolved gases at the reactor outlet (this implies use of gas bubbles in the core system), while in Systems D and E a portion of the fuel solution is contacted with vapor from the gas system in a side stream. This contacting could be accomplished by means of a vapor-liquid jet (using the pressure drop across the main circulating pump to provide the energy for circulation of the liquid and vapor) or by means of a blower for circulating the gas stream. In Systems D and E, use of a holdup tank in the fuel solution line leading to the contactor, can be employed to decrease the deuterium content of that side stream to such a point that explosive mixtures are avoided in the gas phase of the high pressure system. In system C it will be necessary to have negligible volume in the entire gas system as shown in order to avoid the hazard of explosions and to provide a high pressure recombiner before the condenser.

The design equations for the systems shown in Figure 3 are presented in the Appendix and these have been evaluated in a few flow conditions. The results of these calculations are shown in Table I. The systems have been evaluated for condensate production rates of 1.5 gal per minute and in the cases of Systems D and E for the processing of 20 gal/min of fuel solution. The solubility of xenon in the condensate was included in the calculations. It was assumed that a jet eductor was used as a contactor and the gas re-

recirculation rate from the vapor space was calculated using a fuel circulation of 20 gal/min and the jet equation given in CF 56-2-81 with an efficiency of 5%⁽¹⁴⁾. The design equations given in the appendix can be used for evaluating the performance of a system under other conditions of temperature and flow rate. The 20 gal/min circulation rate was selected for these preliminary calculations so that a 20 gal holdup tank would give a one minute holdup time for recombination of deuterium and oxygen in the fuel solution being processed⁽¹⁾. In the final design of such a system, the flow rate would be selected to provide for stripping and condensate production and the holdup volume selected to give the desired amount of recombination.

The results given for System E in Table I show that the xenon poison fraction can be reduced to less than 1% using vapor letdown systems. However, the amount of gas which must be letdown requires a large supply of oxygen to the reactor system and provision for disposal of this large volume of gas. For example, the gases letdown in System E will consist principally of oxygen and will demand a supply of oxygen at a rate of about 2300 liters/min (STP). For System 3C to be operable, a high-pressure recombiner is required so that explosive mixtures would not be present in the condenser.

In comparing Systems C, D and E, System E is considered to be the most compatible with the design criteria of HRE-3. However, removal of the large volume of oxygen from the off-gas stream would probably be required prior to sending the off gases to a charcoal bed; this could be accomplished by introduction of hydrogen and combustion of the oxygen in the off gas system. Considering this process complication and the amount of oxygen necessary at 1500 psi, the reduction of xenon poisoning in HRE-3 by vapor let-down does not appear to be as attractive a method as processing the condensate at high pressure (See Section 3.3).

3.3 Removal of Iodine from Condensate Produced at High Pressure

So long as condensate is required for purging the high pressure pumps, the design criteria set forth for HRE-3 demand that condensate for purging be produced in the high pressure system.

A non-condensable gas, such as oxygen, used to strip xenon and iodine from the fuel solution, could also be employed to produce condensate. Bolger and Maak⁽¹⁾ have reported on a system which circulates oxygen through the reactor pressurizer and then through a condenser to produce condensate. Use of such a system to control xenon poisoning as well as produce condensate offers an opportunity to simplify the overall reactor design.

The inert gas is contacted with fuel solution so that some water vapor, xenon, iodine, deuterium, oxygen and any other volatile components of the fuel solution are transferred into the gas stream. The gases are then passed through a condenser where appreciable iodine will dissolve in the condensate although very little xenon will dissolve. Treatment of the condensate for the removal of iodine thus offers a possibility for the reduction of the xenon poisoning in HRE-3.

Figure 5 shows simple diagrams of three processes which would reduce the xenon poison level in HRE-3 by removal of iodine from condensate at high pressure. Block and line diagrams used for deriving design equations are shown and all the auxiliary streams are not shown. Due to the explosive conditions of the gases which would exist in the gas space of System 5F, where the fuel solution and gas phase are in equilibrium with respect to the concentration of non-condensable gases, System F would be inoperable for HRE-3.

Systems G and H isolate the vapor space from the main fuel solution by means of a vapor liquid contactor, such as a liquid-driven gas eductor so that the concentration of deuterium and oxygen in the core system is not necessarily in equilibrium with the vapor space. Use of a holdup tank in the fuel solution line leading to the contactor provides time for recombination of deuterium and oxygen to avoid the presence of explosive mixtures in the vapor space. Providing the amount of gas bubbles which go directly from the core to the gas space, through the surge line, is not large, this procedure should keep the deuterium concentration in the gas phase at a reasonably low value. Systems G and H differ only in that in System H the off gas from the condenser is recycled through the jet, whereas in System G the gas is taken directly from the vapor space for contacting the fuel solution. Furthermore, the equations derived for System H also allow for the direct passage of gas from the core system into the vapor space to allow for the separation of any gas bubbles which may be present in the circulating core fluid.

The Henry's Law constants which have been reported for iodine in fuel solution and iodine in condensate under reactor temperature and pressure favor the removal of iodine from the fuel solution and the absorption of iodine into a water or condensate stream. (Henry's Law constant, H , for fuel solution at 280°C = 7 mol fraction of iodine in the vapor per mol fraction iodine in the fuel solution, while for iodine in water H = 0.2 mol fraction iodine in the vapor per mol fraction iodine in the liquid water phase^(5,6)).

Stripping of the iodine from the condensate before the condensate is returned to the reactor can be accomplished at either low or high pressure. Considering the design criteria for HRE-3, processing at low pressure would involve dependence upon practically continuous letdown of condensate to limit decay to xenon and at least reasonably frequent pump up of the condensate to a high pressure condensate storage tank. Therefore only removal of iodine from the condensate at high pressure will be discussed in this report. In Figure 5, the details of the iodine removal system have not been shown since there are several methods for removing the iodine from the condensate; these methods will be discussed separately in Section 3.4. In order to simplify the calculations of Systems F, G and H, the condensate returned to the reactor was assumed to contain no xenon or iodine. Actually this condensate will contain a very small amount of xenon (the solubility of xenon in condensate is quite small).

The design equations for these three systems have been derived and are given in the Appendix. A few values for the xenon poison fraction at various processing rates have been calculated and are presented in Table II.

When approximately 1 to 1-1/2 gallons/min of condensate is produced, a xenon poison fraction of the order of 1% can be achieved by System H. The loss of oxygen from the system is much less than in the systems shown in Figure 3. The maximum loss of oxygen in System H is shown in Table II, (assuming that all of the inert gas is oxygen). Use of the condensate as the media for the removal of iodine from the primary high pressure system thus acts as a seal against the removal of oxygen and deuterium from the high pressure system; very little xenon is also removed in the condensate. It is necessary to remove the condensate continuously so that no significant quantity of the iodine absorbed in the condensate decays to xenon prior to removal. The solubility of xenon in the condensate is much less than that of iodine, so that xenon formed by decay of iodine in the condensate will tend to evolve from the condensate.

In all of the systems shown in Figures 3 and 5, the fuel solution returning to the main core system will tend to be saturated in the non-condensable components of the stripping gas stream. In cases where the explosive nature of this stripping gas is controlled so that the gas contains very little deuterium (and hence is practically all oxygen), the fuel solution returning to the core system will be essentially saturated in oxygen. As this returning solution is mixed with the main circulating stream and flows back into the reactor core, where radiolytic deuterium and oxygen are produced, the fuel solution will become supersaturated with gas and gas bubbles will form. The larger the side stream of fuel solution being processed and the more oxygen that this stream brings back into the main core, the greater will be the tendency for gas bubbles to form in the core and to be circulated around the core system. Heating of the processed fuel solution prior to its return to the main core loop will limit the amount of oxygen which the process stream will carry into the main core loop. This heating can be accomplished electrically, although decay heat and heat of recombination will also tend to raise the temperature of the stream, while the evaporation of the D₂O into the inert gas being recycled will tend to lower the temperature of the fuel solution being stripped.

The presence of bubbles circulating in the core will aid in the stripping of iodine and xenon and will also tend to strip deuterium from the fuel solution. Any gas bubbles present at the core outlet can be separated from the fuel solution and fed into the gas space in the pressurizer. Providing that the flow ratio of the deuterium carried into the gas space from the core to the oxygen stripping stream being recycled is properly maintained, explosive conditions should not arise in the gas space. Recombination of the deuterium in this gas will be necessary and can be accomplished by absorption of the deuterium in the gas fed to the jet by the deuterium-free liquid entering the jet from the hold-up tank (See Systems 5G & 5H). By providing adequate time for absorption and recombination in the liquid phase the deuterium content of the gas reentering the pressurized gas space can be maintained below the explosive limit. Development work is required to determine the rate of simultaneous absorption and recombination of the deuterium in the liquid so that the proper method and time of contacting are provided.

3.4 Transfer of Iodine from High Pressure System to Off Gas System

As discussed in the previous sections the xenon-135 poison can be transferred from the high pressure system to the low pressure system in two ways. This poison can be removed as iodine-135 dissolved in a liquid stream such as condensate, or the poison can be removed in a vapor stream as xenon-135. If the poisoning by Xe-135 is to be low, the concentration of xenon in the fuel solution must be low and hence the concentration of xenon in the gas phase in contact with the fuel must be low. Furthermore, xenon has a low solubility in condensate (a high Henry's Law Constant = 73 mol fraction gas/mol fraction liquid), so that removal of xenon in condensate is not practical. Reduction of Xe-135 poisoning by removal of xenon from the main high pressure system requires let-down and make-up of large volumes of gas due to the low tolerable concentration of xenon-135 in the gas (See Table I for gas let-down required).

The amount of oxygen lost from the high pressure system can be limited by taking advantage of the fact that iodine is considerably more soluble in condensate or water at reactor temperature and pressure than it is in fuel solution. Thus as the D_2O is stripped from the fuel solution to produce condensate, iodine is also stripped and as the condensate is condensed some of the iodine will dissolve in the condensate. Table II shows that if 1 to 1-1/2 gal/min of condensate are produced, sufficient iodine will dissolve in this condensate so that the xenon poison fraction in the reactor will be reduced to approximately 1%. Table II shows that for System H, approximately 0.38 cfm (STP) of oxygen will dissolve in condensate and will be transferred along with the iodine.

There are several methods for removing the iodine from the condensate before the condensate is returned to the condensate storage tank and for purging of pumps and concentration control. All of these systems would be expected to operate at high pressure so as to comply with the design criteria that there shall be substantially no need for continuous letdown and pump up. Block and line flowsheets with possible systems are presented in Figures 6, 7 and 8. These systems have not been evaluated in any detail but are presented as concepts for further study.

One of the simplest concepts is shown in Figure 6. In such a process, the condensate produced by stripping of the fuel solution is fed to an evaporator and there completely vaporized. The vapor is passed through a bed packed with silver coated pellets where the iodine would be absorbed and held up for decay to xenon. The vapor from the silver bed passes into the condenser which would produce the condensate for return to the high pressure condensate storage tank. Conversion of the iodine to xenon would change the solubility of the poison by a factor of approximately 350 (H for iodine) in water at $280^{\circ}C = 0.2$ mol fraction of vapor/mol fraction of liquid, while H for xenon in water at $280^{\circ}C = 73$ mol fraction of the vapor/mol fraction of liquid⁽⁵⁾. Thus very little xenon will dissolve in the condensate. The off gas from the condenser could be passed through cold traps, let down to low pressure, and finally fed to charcoal beds for holdup of the radioactive fission product gases. Some oxygen would be present in the feed condensate and would leave the system primarily in non-condensable off-gases. Depending

upon the quantity of this oxygen and the design of the off gas system it might be necessary to remove the oxygen prior to feeding the off gases to the charcoal beds.

Since the design criteria dictate that the system shown in Fig. 6 operate at approximately 1500 lb, the evaporator would operate at approximately 305°C. This requires the use of electrical heating for evaporation. To evaporate 1.5 gal/min of condensate it will be necessary to supply approximately 110 Kw of electrical power. The bulk of this energy is for supplying the latent heat of vaporization and only a small amount goes into heating the condensate to 305°C, since the condensate will leave the main high pressure loop at about 280°C. Complete boiling of the condensate in a single stage evaporator appears to be the most effective means of putting the iodine into the gas phase, since the iodine is the less volatile component in the iodine-water system and would tend to seek the bottom of any distillation or stripping tower. A numerical flowsheet evaluation of this process was carried out in conjunction with System 8. The results are discussed in Section 3.4.

Another method for removing iodine from the condensate that has been suggested is the addition of NaOD into the condensate to convert the iodine to the non-volatile iodide and iodate forms (14). Figure 7 presents a block and line flowsheet of what such a system might look like where the heavy water. NaOD would be pumped into a high pressure storage tank (intermittently (to avoid the need for continual pump up) and then fed into an evaporator. Following fixation of the iodine by the NaOD the heavy water would be boiled off and condensed and returned to the reactor system. Any oxygen, xenon, krypton or other non-condensable gases introduced in the feed condensate, plus any xenon resulting from iodine decay in the evaporator would be fed to an off gas system. A solution of radioactive sodium-iodide and sodium-iodate will be let down into storage tanks for decay to xenon. Since the half life for iodine-135, the xenon-135 producing isotope, is 6.68 hours, a holdup time of approximately 3 days should be sufficient to reduce the iodine content from the condensate to a negligible level. Thus the use of three or four storage tanks each holding approximately 1 day's production of condensate should be sufficient. The volume of condensate letdown from the reactor system will depend upon the concentration at which the iodine is removed from the evaporator; removal of the iodine as 0.001 molar would require approximately 13 gallons/day of letdown from the evaporator. (this implies a makeup rate of approximately 13 gallons/day of sodium hydroxide solution). A means for stripping the xenon in the stored condensate is shown in Figure 7. Following decay of the iodine and stripping of the xenon the sodium hydroxide solution could be recycled to the evaporator. Without detailed evaluation of either system, use of NaOD as shown in Figure 7 appears considerably more complicated than the evaporation system of Fig. 6.

Instead of the use of sodium hydroxide, which might result in cracking or embrittlement at the temperatures and pressures required, a solution of silver sulfate could be added to the evaporator and silver iodide removed by hydroclone for letdown to decay. Development work would be required to determine whether the silver iodide would remain suspended in the solution for removal by hydroclone or whether it would precipitate out on the surfaces

of the evaporator. A conceptual flowsheet for such a system is shown in Figure 8-1.

Also shown in Figure 8-2 is a concept in which the need for evaporation is eliminated. Here silver sulfate is mixed with the condensate feed in near-stoichiometric ratio and the resulting silver iodide removed in a hydroclone, providing that the proper crystal size can be produced. Data are required on the solubility of silver iodide in condensate at reactor temperature to determine whether the removal of iodine is sufficiently great and whether the introduction of silver would constitute a significant nuclear poison when the condensate was returned to the core system.

Data on the solubility of silver iodide in fuel solution are not available, but using the handbook solubility of silver iodide in hot water as a very rough measure of the possible solubility of AgI in fuel solution, the poisoning by the dissolved silver would be negligible (the silver concentration under such an assumption would constitute approximately 0.001% nuclear poison). Introduction of some silver in the condensate might, however, cause precipitation of silver iodide in the reactor resulting in coating out of silver iodide in the reactor and on heat transfer surfaces.

The same concept could be employed in the addition of silver sulfate directly to the fuel solution (probably in a side stream containing hydroclones and continuous filters) providing there were no undesirable reaction between the silver and the fuel solution, and assuming that the solubility of silver iodide in fuel solution is approximately that in hot water. Use of direct contact of silver solution with the condensate or fuel solution would require considerable chemical exploratory work, but successful application of silver sulfate to either the condensate (without subsequent evaporation) or the fuel solution itself would result in considerable simplification of the xenon removal system, and it is recommended that the Chemical Technology Division be requested to carry out a brief evaluation of this technique.

Use of ion exchange resins was also considered as a means of removing iodine from the condensate (after conversion of iodine to an ionic form). The relatively poor thermal and radiation stability of the resins plus the need for cycling of parallel beds to allow for decay were considered to make such a process impractical.

3.5 Evaluation of Combined Oxygen Pressurization-Condensate Production System

With the information now available, use of the systems of the type shown in Figure 5-H and Figure 6 appears to offer the best opportunity of combining condensate production at high pressure with reduction of xenon poisoning in the reactor. Figure 9 presents a block and line flow sheet of such a system. Several assumptions were required in calculating the flow sheet quantities, and development work is required to check on these assumptions and to provide additional data required for design and a more complete appraisal of the flowsheet.

The values given in Table II for this system are based on a fuel solution processing rate of 20 gal/min. At a condensate production and treatment rate of 1.5 gal/min, the resulting xenon poison level should be approximately 1%.

In this system a holdup tank of 20 gallons is provided in the line feeding the fuel solution into the processing system in order to provide for recombination of deuterium and oxygen. It is assumed that the deuterium concentration in the liquid entering the jet is essentially zero. The volume of gas bubbles rising directly from the core to the pressurizer will be a function of the temperature at which the stripped fuel solution is returned from the pressurizer into the circulating line (since this determines the concentration of oxygen in the fuel solution leaving the pressurizer). The values of Table II reported are for a liquid being returned at 280°C. This is approximately the temperature which is calculated by heat balance around the system allowing for decay heat, recombination heat in the fuel solution and loss of sensible heat which goes into heat of vaporization of the condensate produced. The quantity of bubbles introduced into the main circulating loop is dependent upon the flow rate of the fuel solution being processed and the temperature in the vapor space and of the liquid returning from the pressurizer. A holdup tank in the fuel solution line leading to the pressurizer is provided to allow one minute holdup so that essentially all the deuterium in the fuel solution will recombine.

In calculation of the deuterium concentrations for the system shown on Fig. 9, it was assumed that the deuterium introduced into the pressurizer by the gas bubbles from the core goes immediately to the vapor space and the deuterium concentration is there diluted by the recirculating gas stream (this assumption introduces maximum deuterium into the gas phase by not allowing for absorption and recombination in the pressurizer). It was assumed that the deuterium in the gas stream leaving the condenser was completely absorbed and recombined in the jet and the line leading from the jet to the pressurizer. A check of the validity of this assumption can be obtained by assuming that the rate of absorption in the jet and pipe is infinite so that the deuterium concentration in the gas and liquid in the jet and the line following it are at all times in equilibrium. From this assumption and the rate expressions for recombination at a given copper concentration it would be possible to calculate the contact time (therefore the length of line for given flowrate) required to absorb and recombine any given percentage of deuterium in the gas stream entering the jet. From a knowledge of the volume of gas leaving the core and its concentration of deuterium, one could calculate the gas composition required in the vapors entering the recombiner from the jet recombiner line in order to produce a given concentration of deuterium of the mixed gas in the pressurizer space (this latter value would be based upon the assumed safe lower explosive limit for deuterium and oxygen). The composition of the gas entering the jet, the gas composition leaving the contact zone (jet and pipe line connecting the jet and the pressurizer), and the time required to achieve the change in composition would thus all be determined for a given set of gas and liquid flow rates, copper concentration, and reactor operating conditions. The length of the pipe necessary to provide the contact time required would thus be fixed. If a practical length of pipe resulted, the operating conditions selected could

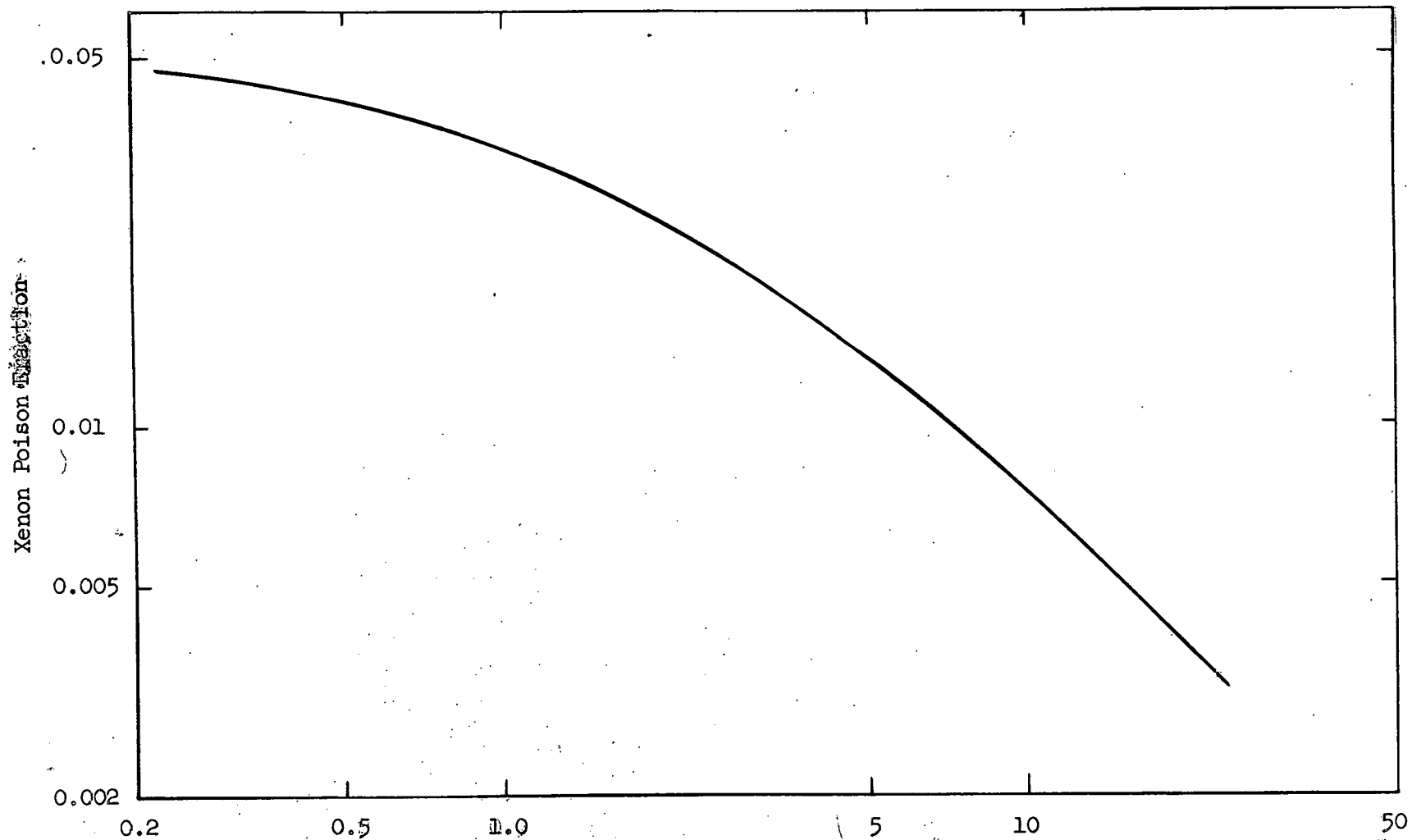
be considered safe. The calculations should be corrected when data can be obtained upon the rate of absorption of the deuterium into the fuel solution in the jet and pipe.

Approximately 110 kw of electrical heat must be supplied in order to vaporize the condensate in the evaporator at the required temperature of about 305°C and 1500 lb pressure. The bulk of this heat is for heat of vaporization. This is a considerable electrical energy input and may impose a design problem. Furthermore, in order for the system shown in Figure I to operate, the silver bed must operate close to the disassociation temperature for silver iodide. Since the evaporator must operate at a temperature of about 305°C and the silver iodide is reported to begin to decompose at temperatures of 350 to 400°, the decay heat of the gases absorbed in the silver may cause the temperature of the silver iodide to rise to a region where decomposition would occur. Should the silver bed be, or become, inefficient in removing iodine, the amount of condensate produced must be increased. If cooling of the silver bed is difficult, the system shown in Figure 6 would be inferior to those employing liquid phase fixation agents, such as shown in Figure 7 or 8.

The jet equation given in report CF 56-2-81⁽¹⁴⁾ was employed in calculating the quantity of vapor pumped by the 20 gal/min of liquid. A jet efficiency of 5%⁽¹⁴⁾ was assumed. It will be necessary in the final design of this system to determine more accurately the relationship between the liquid flow into the jet and the gas pumped.

Fig. 1 Effect of Liquid Letdown on Xenon Poison Level - HRE-3 Core

50 Mw
Core Outlet at 280°C
Volume High Pressure System = 4000 liters
Total Pressure = 1500 psia



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Fig. 1 Effect of Liquid Letdown on Xenon Poison Level - HRE-3 Core

50 Mw
Core Outlet at 280°C
Volume High Pressure System = 4000 liters
Total Pressure = 1500 psia

Fig. 1
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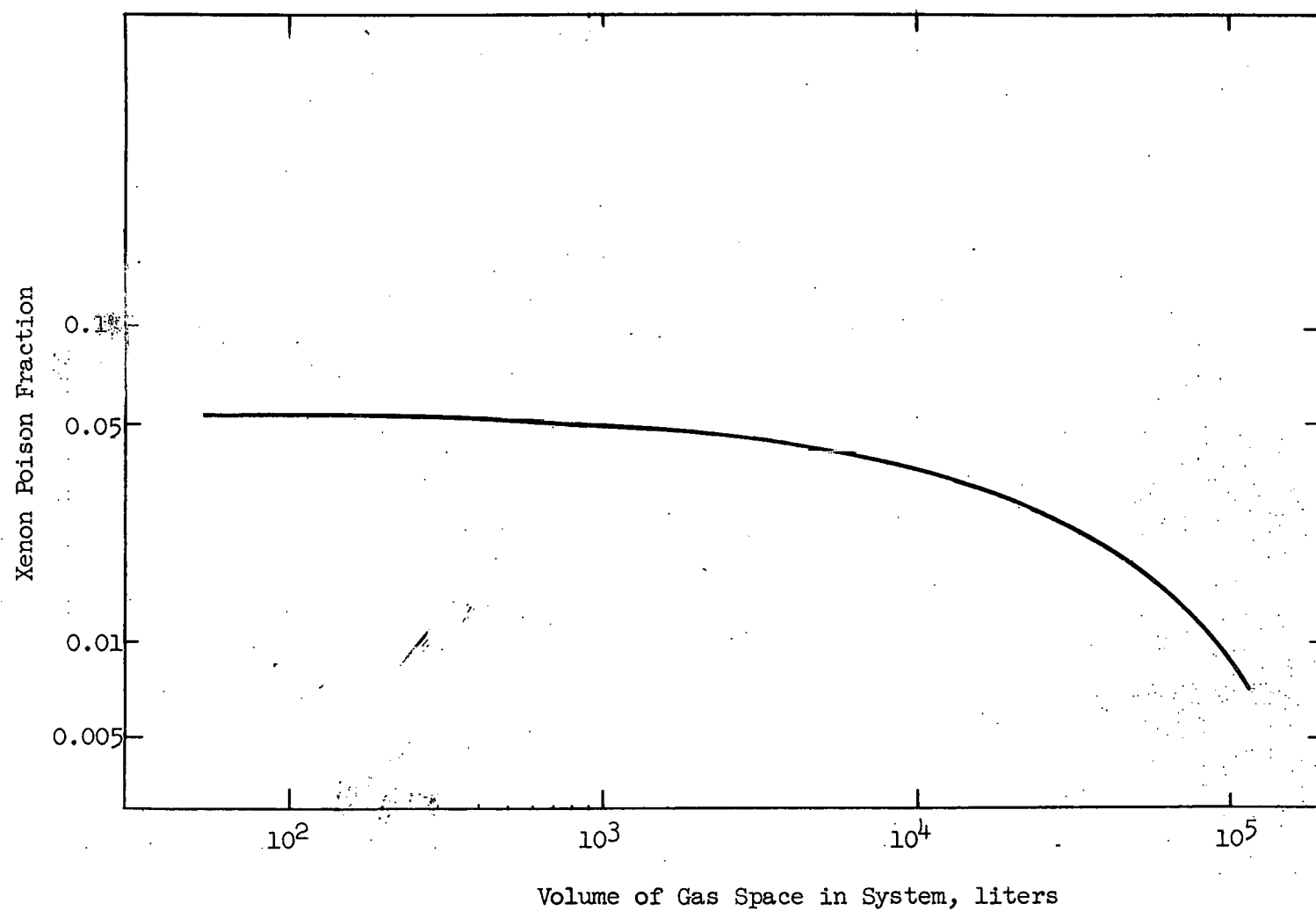
Fig. 2 Effect of Gas Volume on Xenon-135 Poison Fraction in HRE-3 Core System- No Removal of I-135 or Xe-135

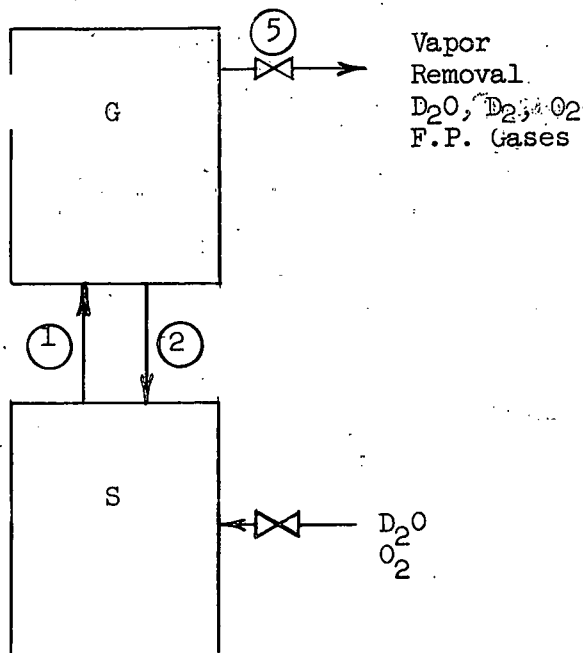
P = 50 Mw

Volume liquid in core system = 4000 liters

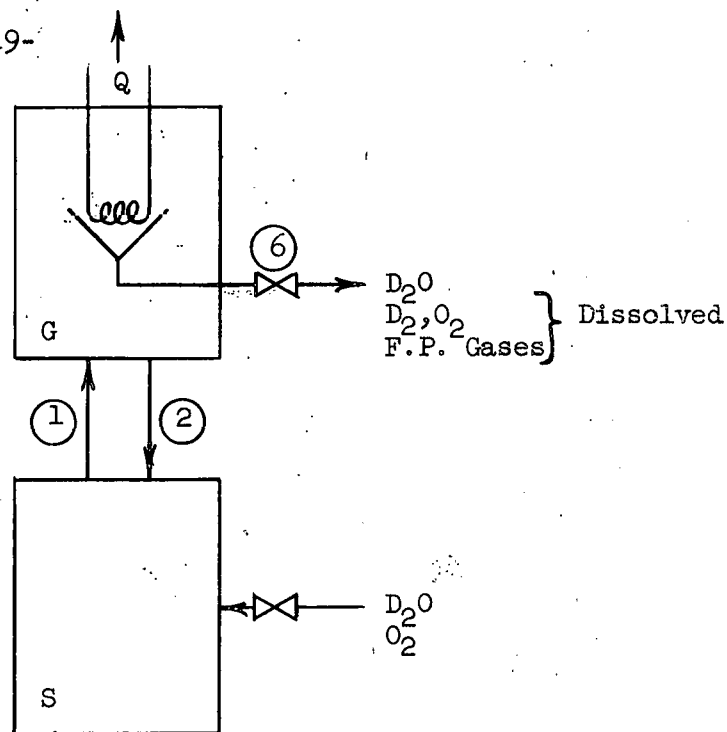
Total pressure at core outlet = 1500 psia

Temperature at core outlet = 280°C





System A
Vapor Letdown



System B
Equilibrium Condensate
Letdown

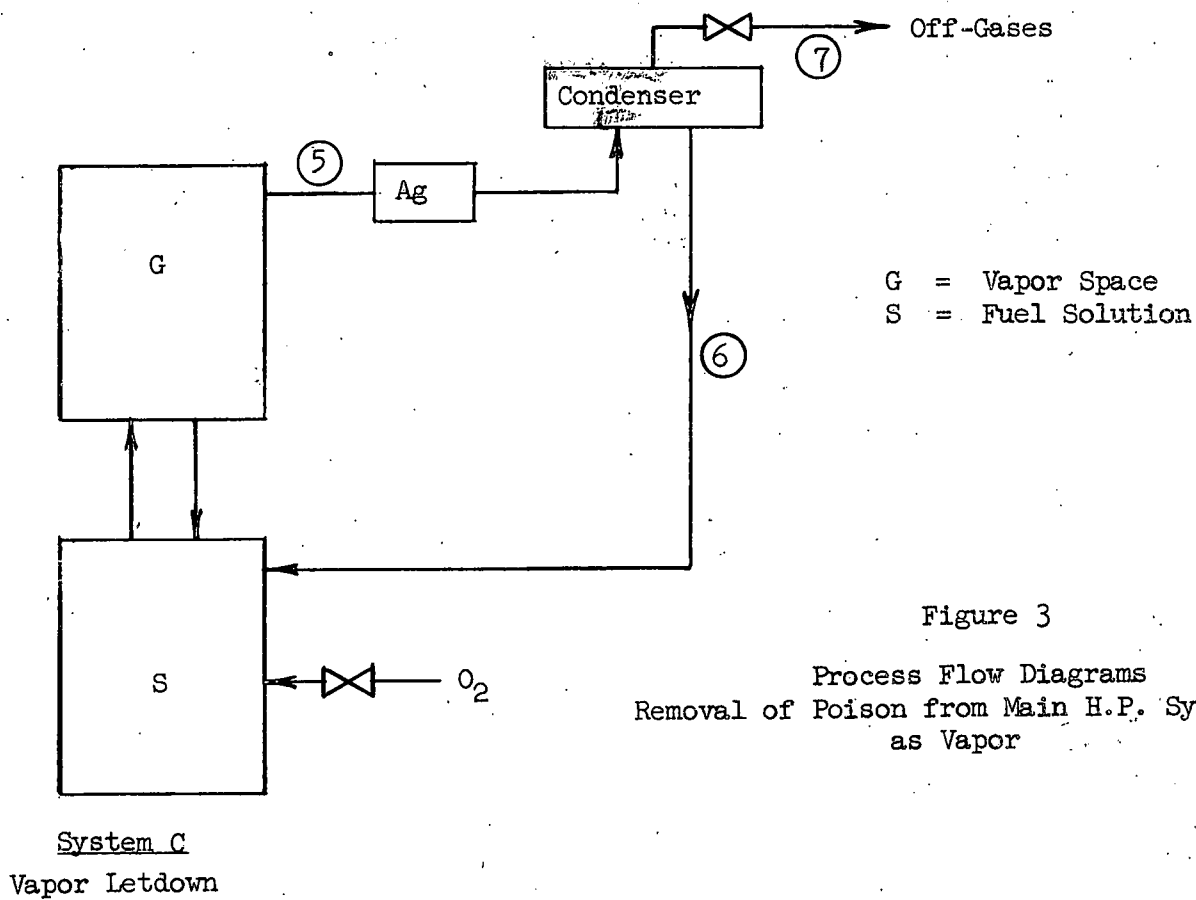


Figure 3
Process Flow Diagrams
Removal of Poison from Main H.P. System
as Vapor

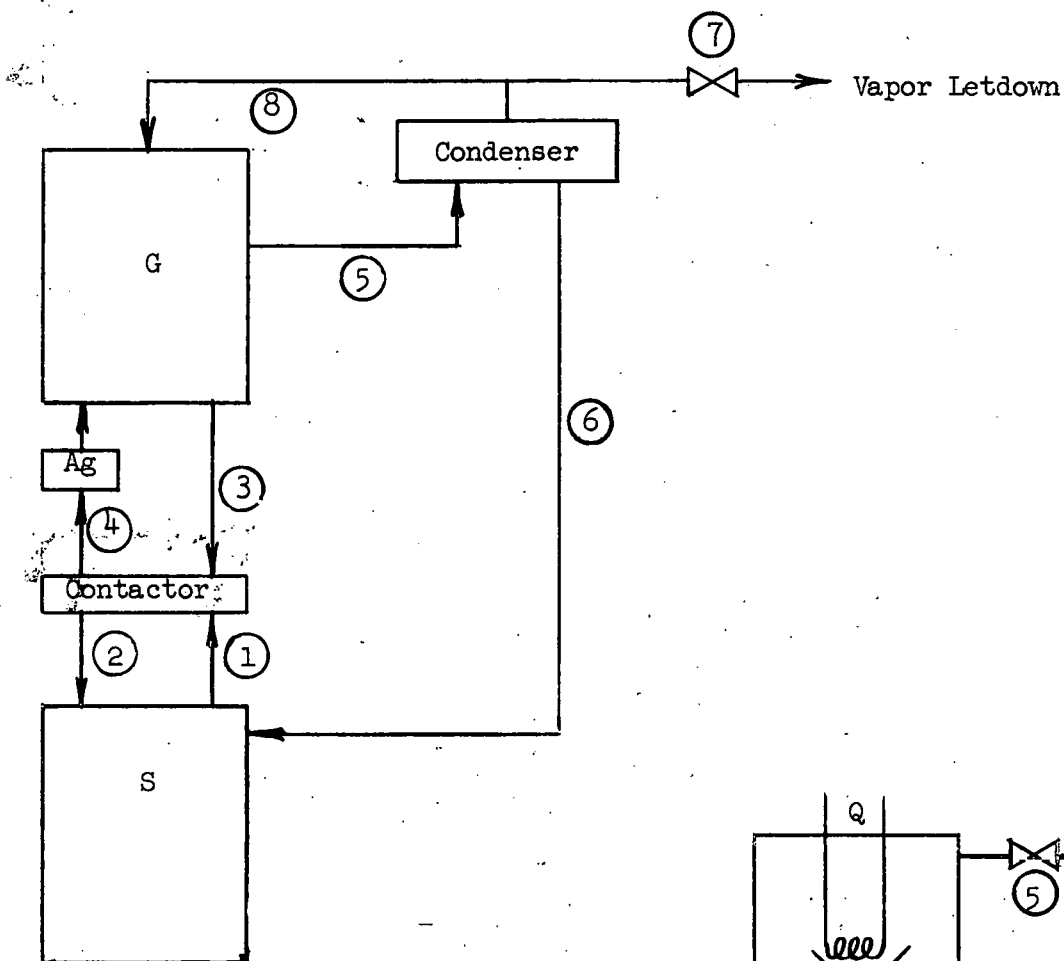
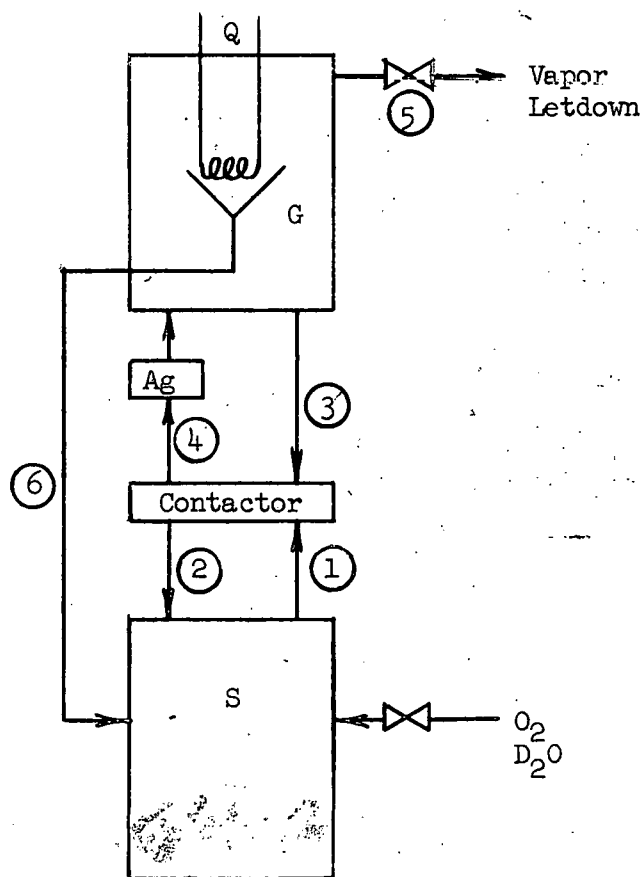


Figure 3 (continued)



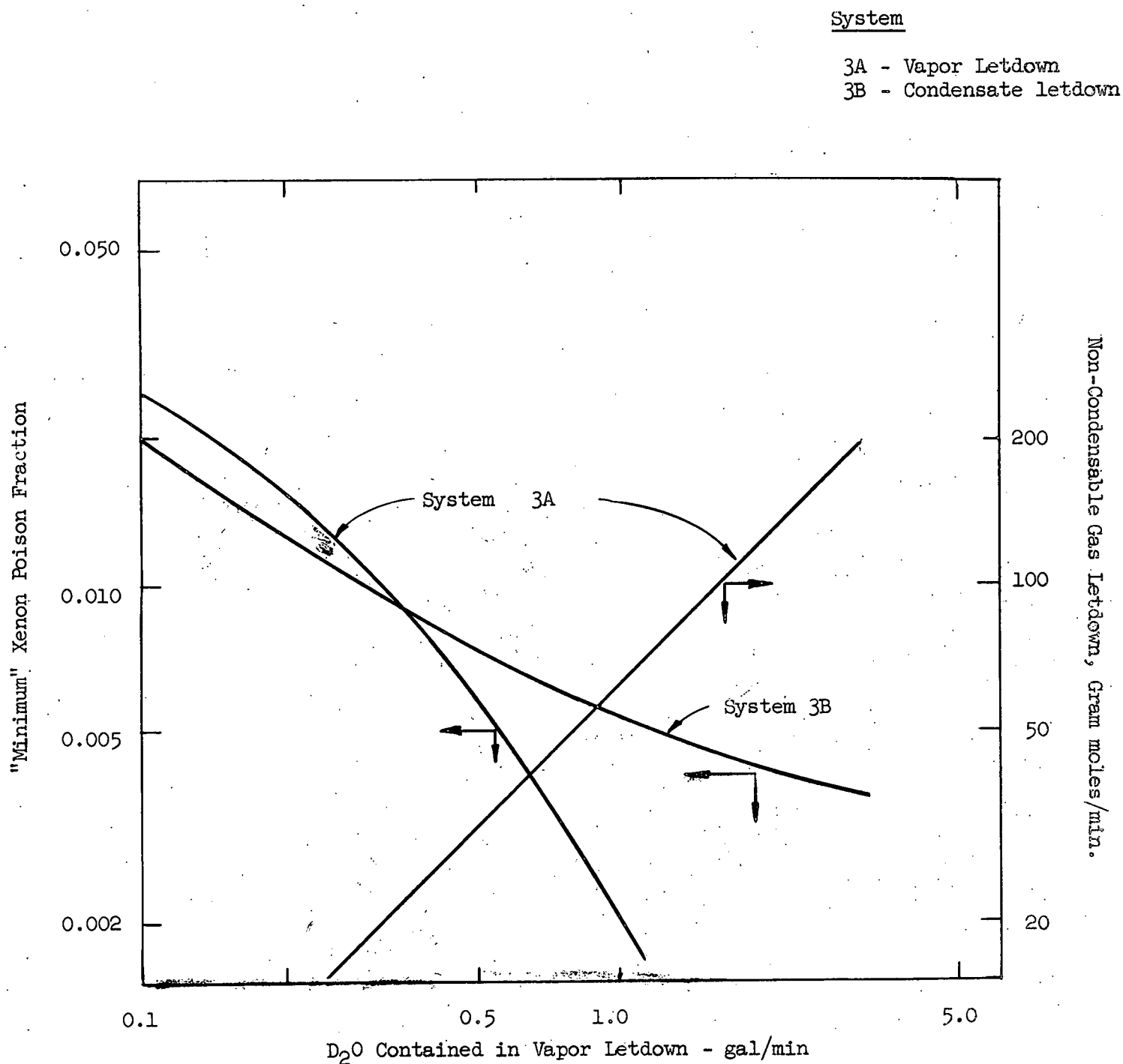


Figure 4. Effect of Direct Letdown of Vapor or Condensate on Xenon Poison Level in HRE-3 Core

50 Mw

Core Outlet Temperature = 280°C

Volume Liquid in High Pressure System = 4000 liters

Total Pressure at Core Outlet = 1500 psia

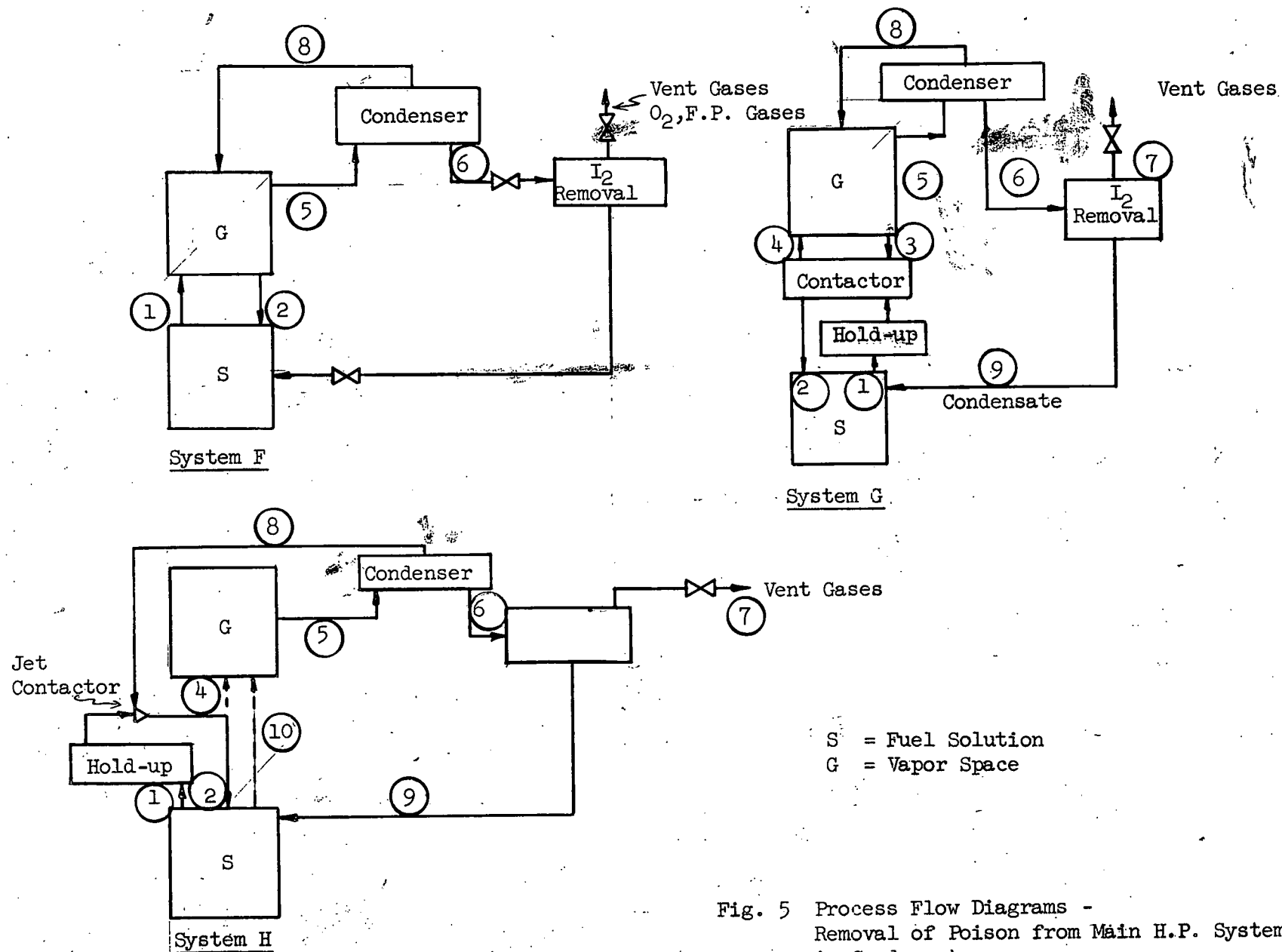
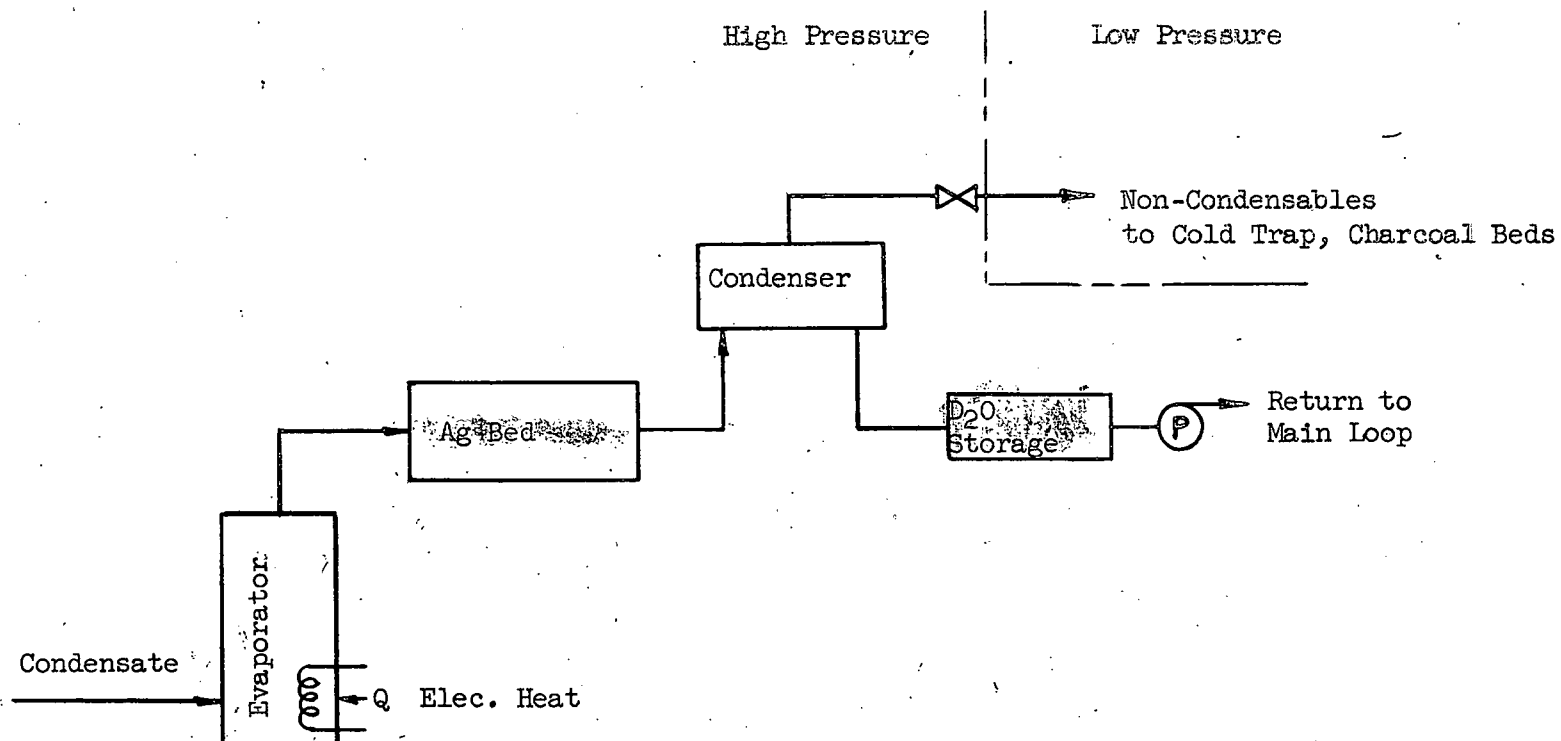


Fig. 5 Process Flow Diagrams -
Removal of Poison from Main H.P. System
in Condensate



Problems:

Heat input to evaporator

Efficiency of Ag bed

Temp. Ag bed. AgI decomposes $\sim 350^{\circ}\text{C}$

Steam at $\sim 315^{\circ}\text{C}$ plus decay heat

Figure 6. Removal of Iodine from Condensate By Conversion to Xenon

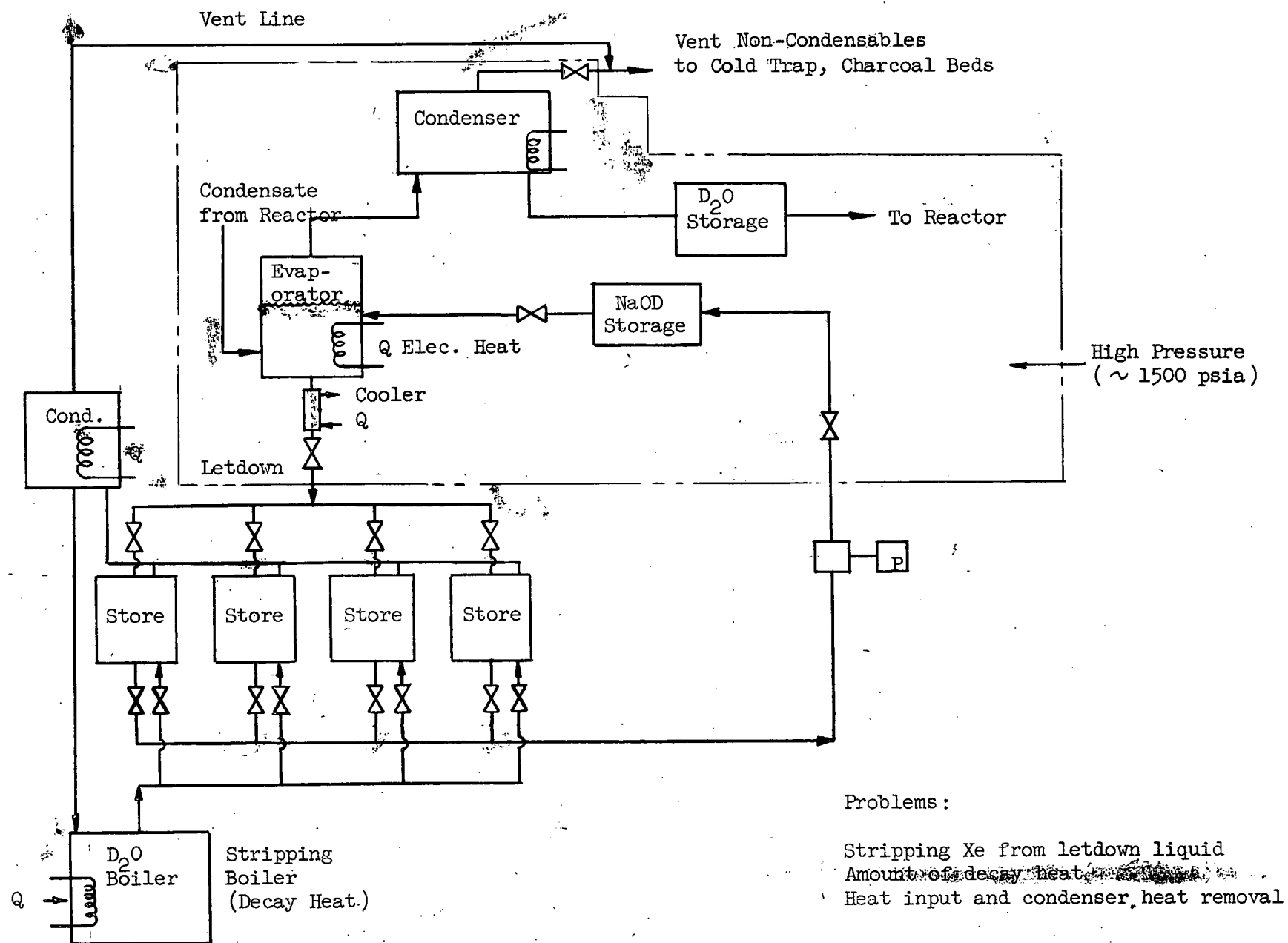


Fig. 7 Removal of Iodine from Condensate By Chemical Reduction of Volatility

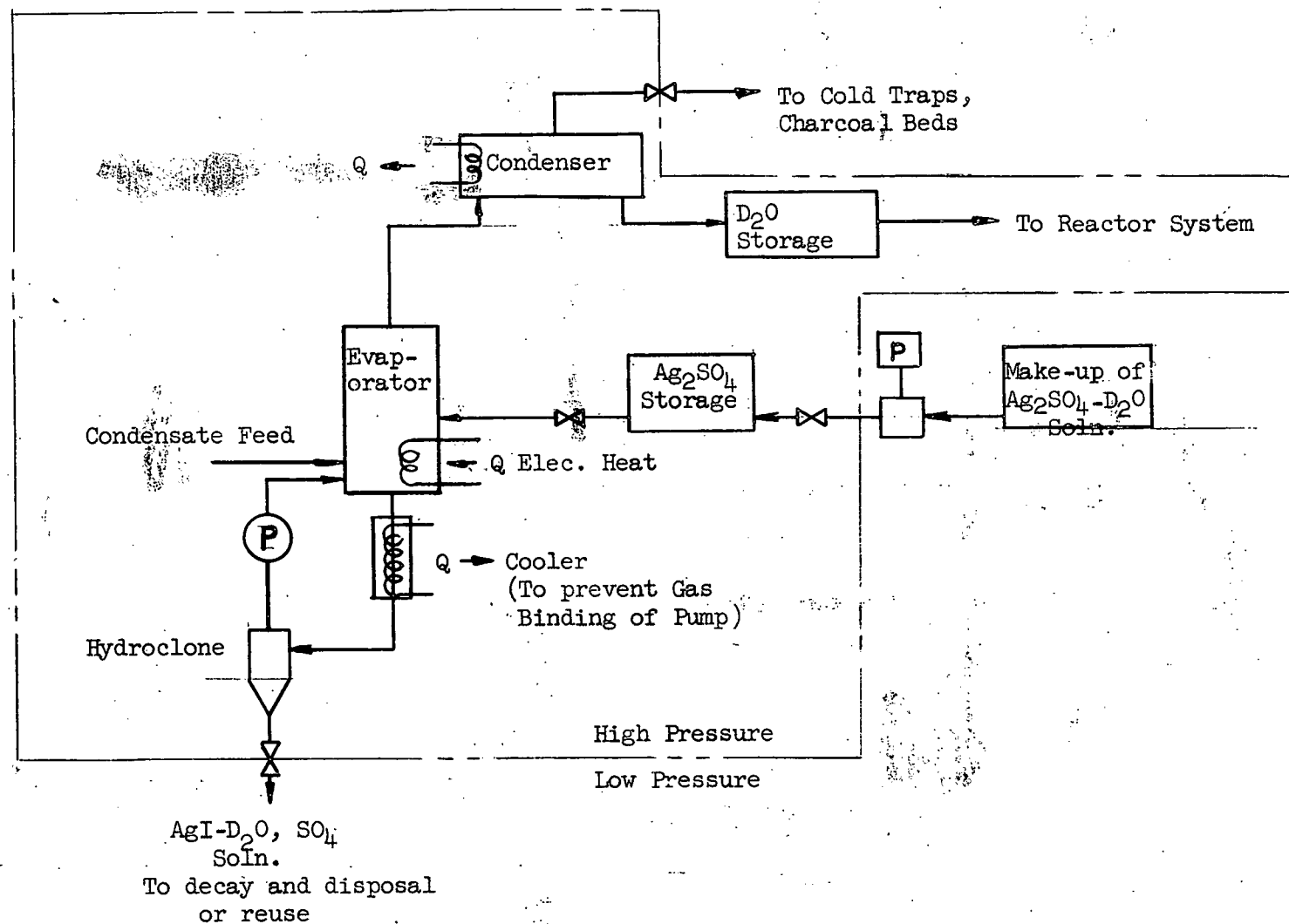


Fig. 8-1 Removal of Iodine from Condensate by Chemical Precipitation

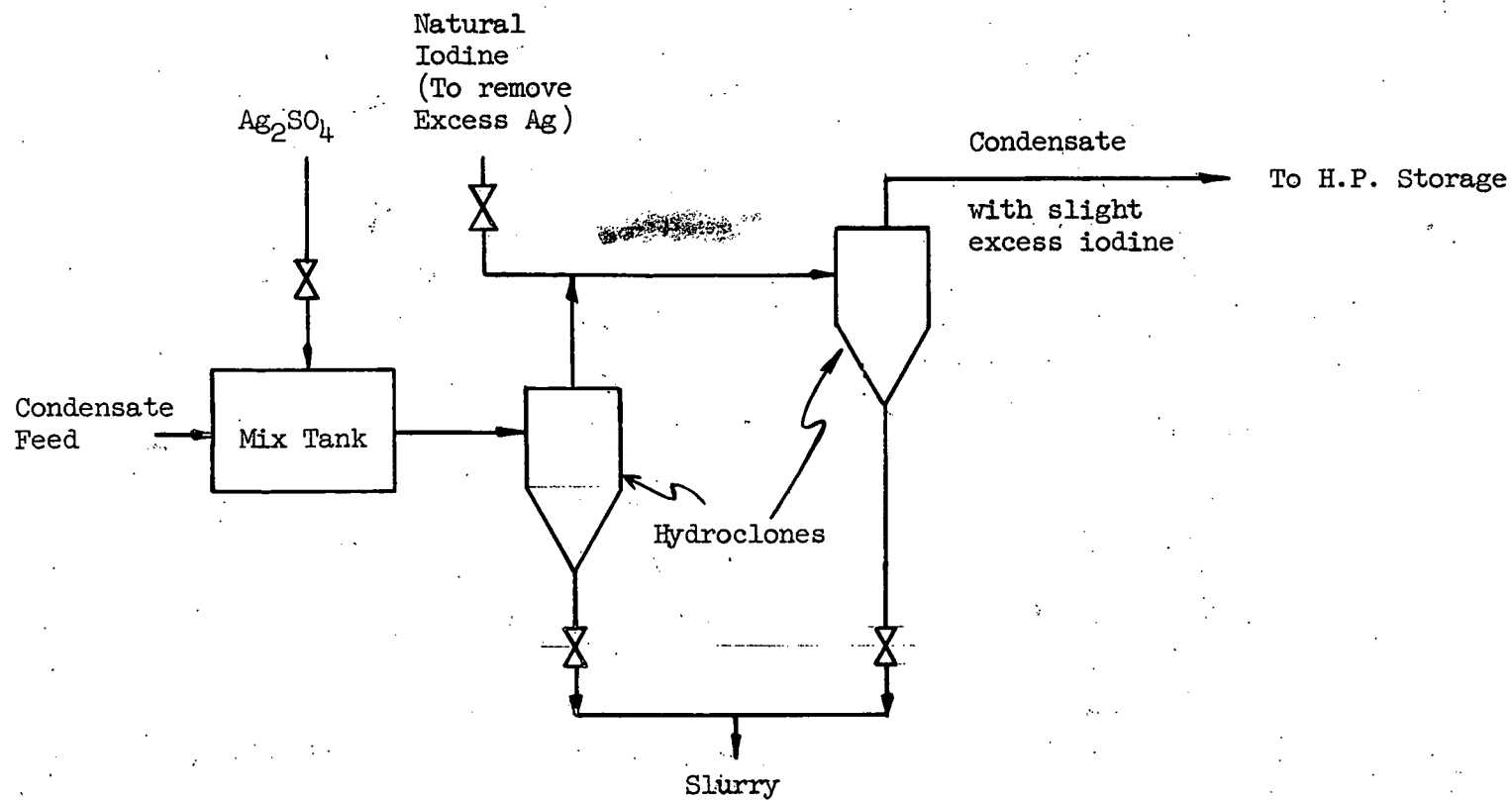


Fig. 8-2 Removal of Iodine from Condensate by Chemical Precipitation

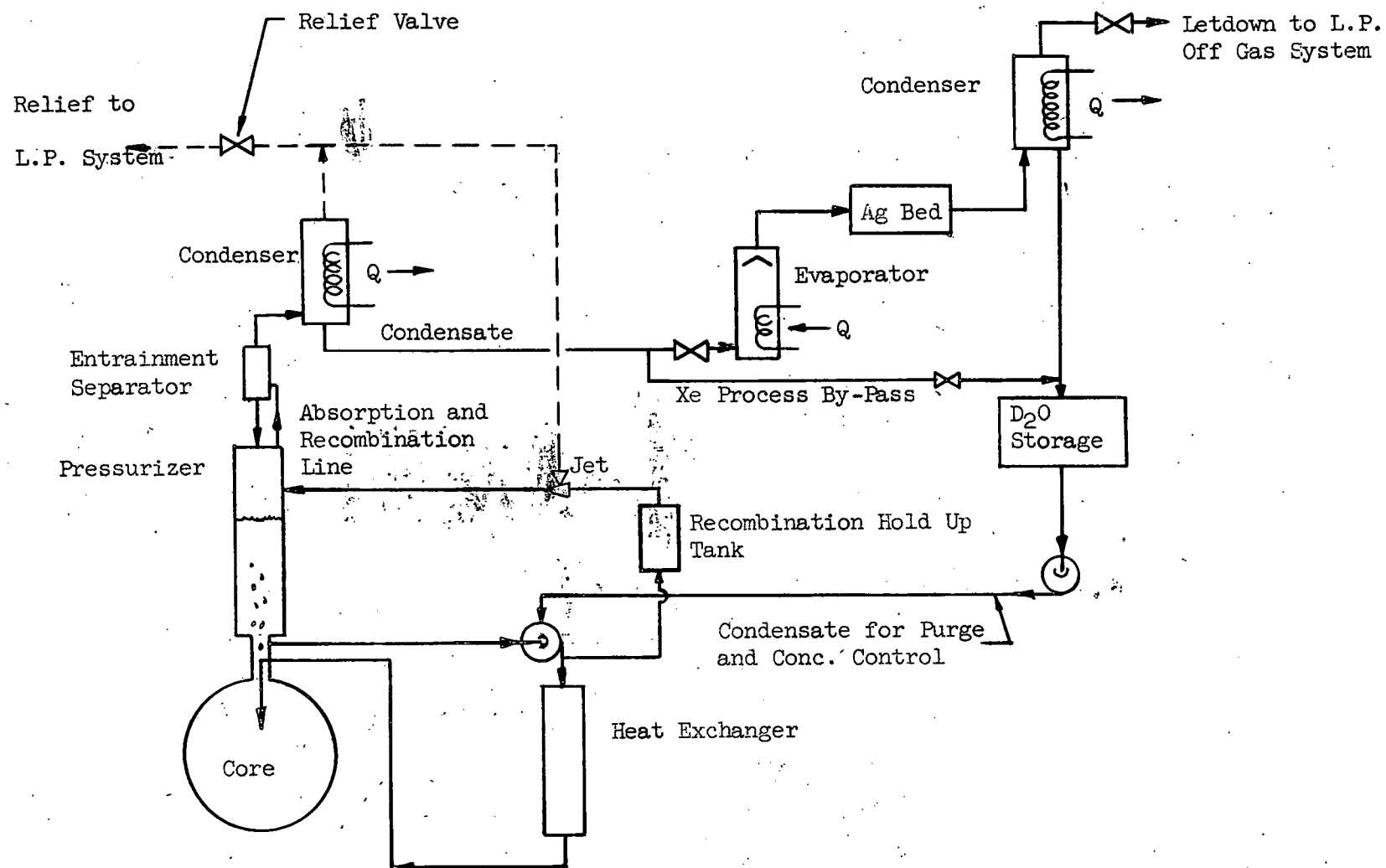


Fig. 9 Combined Oxygen Pressurization, Condensate Production and Xenon Removal System

4.0 Appendix

4.1 Derivation of Design Equations

4.1.1 Systems Containing Gas and Liquid Zones, Each Well Mixed and in Equilibrium Gas Letdown

Iodine Balance:

$$Y_{I\alpha P} = (N_I)_S V_S \lambda_I + (N_I)_G [V_G \lambda_I + (R_I)_G] \quad (1)$$

(See Section 4.3 for nomenclature)

Xenon Balance:

$$Y_{Xe\alpha P} + (N_I)_S V_S \lambda_I + (N_I)_G V_G \lambda_I = (N_{Xe})_S V_S [\lambda_{Xe} + \sigma_a(Xe) \phi] + (N_{Xe})_G [V_G \lambda_{Xe} + (R_{Xe})_G] \quad (2)$$

Combining (1) and (2)

$$(Y_I + Y_{Xe}) \alpha P = (N_{Xe})_S V_S [\lambda_{Xe} + \sigma_a(Xe) \phi] + (N_{Xe})_G V_G \lambda_{Xe} + (N_I)_G (R_I)_G + (N_{Xe})_G (R_{Xe})_G \quad (3)$$

Defining f_{Xe} :

$$(N_{Xe})_S = \frac{f_{Xe} \Sigma_f(V)}{\sigma_a(Xe)} \quad (4)$$

For equilibrium between fuel solution (S) and vapor (G)

$$(N_{Xe})_G = \frac{(MW)_S \pi (K_{Xe})_S (N_{Xe})_S}{\rho_S Z_a RT_G} = \theta_S (K_{Xe})_S (N_{Xe})_S \quad (5)$$

$$(N_I)_G = \frac{(MW)_S \pi (K_I)_S (N_I)_S}{\rho_S Z_a RT_G} = \theta (K_I)_S (N_I)_S \quad (6)$$

Combining (1) and (6)

$$(N_I)_G = \frac{\theta_S (K_I)_S Y_{I\alpha P}}{V_S \lambda_I + K_I \theta [V_G \lambda_I + (R_I)_S]} \quad (7)$$

Combining (3), (5) and (7), get general expression

$$(N_{Xe})_S = (Y_I + Y_{Xe})\alpha_P - \frac{\theta_S (K_I)_S (R_I)_G Y_I \alpha_P}{V_S \lambda_I + (K_I)_S \theta_S [V_G \lambda_I + (R_I)_G]} \quad (8)$$

$$\frac{V_S [\lambda_{Xe} + \sigma_a(Xe)\phi] + \theta_S (K_{Xe})_S [V_G \lambda_{Xe} + (R_{Xe})_G]}{}$$

For System A (See Figure 3-A)

$$(N_I)_G Q = (N_I)_G (R_I)_G = \text{atoms I removed/sec} \quad (9)$$

$$Q = (R_I)_G = (R_{Xe})_G \quad (10)$$

Substitution of (10) into (8) and use of (4) permits calculation of f_{Xe} as a function of Q for given reactor operating conditions for System 3-A. Amount of D_2O in Q is determined from:

$$Q = \frac{L_c \rho_c (Z_a)_G RT_G (x_{D_2O})_c}{(MW)_c \pi (y_{D_2O})_G} = \frac{L (x_{D_2O})_L}{\theta_L (y_{D_2O})_G} \quad (11)$$

Use of (11), (8), (10) and (4) with the values shown in Sections 4.2 and 4.3 resulted in the values for System 3-A for Figure 4.

For System 3-B - condensate letdown

$$(N_I)_G (R_I)_G = C (N_I)_c = C \frac{(N_I)_G}{\theta_L (K_I)_c} \quad (12)$$

$(N_I)_G$ and $(N_I)_L$ assumed to be in equilibrium

$$(R_I)_G = \frac{C}{\theta_L (K_I)_c} \quad (13)$$

$$(R_{Xe})_G = \frac{C}{\theta_L (K_{Xe})_c} \quad (14)$$

Substitution of (13) and (14) into (8) and evaluation gave values for System 3-B in Figure 4.

4.12 System 3-D

Condensate and vapor in equilibrium; contact between vapor and fuel solution limited and so not in equilibrium; conversion of iodine to xenon; removal of poison from main high pressure system by vapor letdown.

Numbers refer to Figure 3-D.

From a Xe-135 and I-135 balance around gas space through lines 3, 4, 5, and 6, and the assumed equilibrium relationship

$$(N_{Xe})_6 = \frac{(N_{Xe})_G}{\theta_c (K_{Xe})_c} \quad (15)$$

we get

$$(N_{Xe})_G = \frac{Q_4 [(N_{Xe})_4 + (N_I)_4]}{\gamma} \quad (16)$$

where

$$\gamma = Q_5 + V_G \lambda_{Xe} + Q_3 + \frac{L_6}{\theta_c (K_{Xe})_c} \quad (17)$$

A xenon and iodine balance around the liquid system through lines 1, 2, and 6 gives:

$$\begin{aligned} (Y_I + Y_{Xe})\alpha P = & (N_{Xe})_S V_S [\lambda_{Xe} + \sigma_a (Xe)\phi] + L_1 (N_{Xe})_S + L_1 (N_I)_S \\ & - L_2 (N_{Xe})_2 - L_2 (N_I)_2 - L_6 (N_{Xe})_6 \end{aligned} \quad (18)$$

An iodine balance around the contactor gives

$$Q_4 (N_I)_4 = L_1 (N_I)_S - L_2 (N_I)_2 \quad (19)$$

since $(N_I)_3$ is zero due to presence of silver bed.

Assuming equilibrium reached at exit of contactor

$$(N_{Xe})_4 = \theta_S (K_{Xe})_S (N_{Xe})_2 \quad (20)$$

$$(N_I)_4 = \theta (K_I)_S (N_I)_2 \quad (21)$$

Combining (16), (18), (19), (20), and (21)

$$(N_{Xe})_S = \frac{(Y_I + Y_{Xe})\alpha_P + \left\{ \frac{L_2 Q_3}{\theta_S(K_{Xe})_S \gamma A} + \frac{L_6}{\theta_c(K_{Xe})_c \gamma} \left[\frac{Q_4 Q_3}{\gamma A} + 1 \right] - 1 \right\} Q_{44} (N_I)_4}{V_S \left[\lambda_{Xe} + \sigma_a(Xe) \phi \right] + L_1 \left[1 - \frac{L_2}{\theta_S(K_{Xe})_S A} - \frac{L_6 Q_{44}}{\theta_c(K_{Xe})_c \gamma A} \right]} \quad (22)$$

$$\text{and } (N_I)_4 = \frac{Y_I \alpha_P}{V_S \lambda_I \left[Q_4 \theta_S(K_I)_S + L_2 \right] + Q_{44}} \quad (23)$$

$$L_1 \theta_S(K_I)_S$$

where γ is defined by Equation (17), and

$$A = \frac{L_2}{\theta_S(K_{Xe})_S} + Q_{44} \left[1 - \frac{Q_3}{\gamma} \right] \quad (24)$$

Substitution into (23) and (24), and use of (4) permits calculation of the effect of various parameters on f_{Xe} , the xenon-135 poison fraction.

4.13 System 3-E

Similar to System 3-D except external condenser employed and vapor letdown from condenser non-condensables. This means that condensate is not in equilibrium with main vapor space and vapors letdown contain very little D_2O .

Overall balance around condenser:

$$Q_8 + Q_7 = \frac{(Z_a)_7 RT_7}{\pi} \left[\frac{Q_5 \pi}{(Z_a)_G RT_G} - \frac{L_6 p_6}{(MW)_6} \right] \quad (25)$$

A xenon-135 balance around the condenser gives

$$(N_{Xe})_7 = \left[Q_5 - \frac{L_6}{\theta_c(K_{Xe})_c} \right] \frac{(N_{Xe})_G}{Q_8 + Q_7} \quad (26)$$

A xenon and iodine balance through line 3, 4, 6 and 7 gives

$$(N_{Xe})_G = \frac{Q_{44} \left[(N_{Xe})_4 + (N_I)_4 \right]}{\gamma'} \quad (27)$$

$$\text{where } \gamma' = \lambda_{Xe} V_G + Q_3 + \frac{L_6}{\theta_c (K_{Xe})_c} + \frac{Q_7}{Q_7 + Q_8} \left[Q_5 - \frac{L_6}{\theta_c (K_{Xe})_c} \right] \quad (28)$$

$(N_{Xe})_S$ is evaluated using Equations (22) and 23) substituting γ' from Equation (43) for γ in Equation (22).

4.13 System 5-G (See Figure 5)

Removal of nuclear poison from main high pressure loop as iodine-135 dissolved in condensate; removal of iodine from condensate at high pressure; limited contact between high pressure vapor and fuel solution in a contactor; hold-up of fuel solution before stripping to allow for D_2 and O_2 recombination.

Xe-135 and I-135 balance around whole system:

$$(Y_I + Y_{Xe})\alpha P = (N_{Xe})_S V_S \left[\lambda_{Xe} + \sigma_a(Xe)\phi \right] + (N_{Xe})_G \left[V_G \lambda_{Xe} + (R_{Xe})_G \right] + (N_I)_G (R_I)_G \quad (29)$$

Assuming iodine and xenon removed from condensate such that

$$(N_I)_G \approx 0 \text{ and } (N_{Xe})_G \approx 0$$

$$(R_I)_G = \frac{L_6 (N_I)_6}{(N_I)_G} \quad (30)$$

and

$$(R_{Xe})_G = \frac{L_6 (N_{Xe})_6}{(N_{Xe})_G} \quad (31)$$

Xenon and iodine absorbed in condenser. Use countercurrent (reflux) condenser with vapors from pressurizer entering at bottom, condensate leaving at bottom, and non-condensables leaving at top (shown diagrammatically in Figure 5-G). This will produce maximum concentration of iodine and xenon in condensate, and hence minimum condensate requirement for a given rate of xenon and iodine removal (thus for a given f_{Xe}).

For given Henry's Law constants, absorption of xenon and iodine will depend upon the number of equilibrium contact stages provided in the condenser-absorber. The slope of the operating time is $\frac{M}{G}$ and the slope of the equilibrium line is K , when

$$\begin{aligned} M &= \text{moles liquid/sec} \\ G &= \text{moles gas/sec} \\ K &= \text{Henry's Law constant, } \frac{\text{mole fraction in gas}}{\text{mole fraction in liquid}} \end{aligned}$$

For constant K , the number of stages for a given absorption depends on the ratio GK/M . In absorption, for values of GK/M greater than unity (more volatile components) the gas and liquid tend toward equilibrium compositions at the rich end (lower end) of the absorber, while for values of GK/M less than unity (less volatile components), the gas and liquid tend toward

equilibrium compositions at the lean end (upper end) of the absorber. In stripping, the reverse is true; that is, the more volatile components tend to pinch at the lean end of the absorber.

The number of equilibrium absorption stages required can be estimated using the Kremser equation (16). Absorption of 90% of the iodine entering the bottom of the condenser-absorber was assumed desired (Where a jet of the type mentioned in CF-56-2-81 is used with a liquid flow of 20 gpm, KG/M = 0.55 and three theoretical stages would be required for 90% absorption).

$$(R_I)_G (N_I)_G = 0.9 Q_5 (N_I)_G$$

$$(R_I)_G = 0.9 Q_5 \quad (32)$$

Since xenon is much more volatile (insoluble than iodine), from (31) and (15)

$$(R_{Xe})_G = \frac{L_6}{\theta_L (K_{Xe})_L} \quad (33)$$

Iodine balance around mixer through lines 1, 2, 3 and 4

$$L_1 (N_I)_S + Q_3 (N_I)_G = \left[\frac{L_2}{\theta_S (K_I)_S} + Q_4 \right] (N_I)_4 \quad (34)$$

Iodine balance through lines 3, 4 and 6

$$Q_4 (N_I)_4 = [Q_3 + V_G \lambda_I + 0.9 Q_5] (N_I)_G \quad (35)$$

Iodine balance around reactor through lines 1, 2, and 9

$$Y_{I\alpha P} = (N_I)_S V_S \lambda_I + L_1 (N_I)_S - L_2 (N_I)_2 \quad (36)$$

Combining (34), (35) and (36)

$$(N_I)_G = \frac{Y_{I\alpha P}}{\left(\frac{V_S \lambda_I + L_1}{L_1} \right) (AB - Q_3) - \frac{B L_2}{\theta_S (K_I)_S}} \quad (37)$$

where

$$A = \frac{L_2}{\theta_S (K_I)_S} + Q_4 \quad (38)$$

$$B = \frac{Q_3 + V_G \lambda_I + 0.9 Q_5}{Q_4} \quad (39)$$

Similarly combining xenon balance around mixer through lines 1, 2, 3, and 4 plus xenon balance through lines 3, 4 and 6 with Equation (39)

$$(N_{Xe})_S = \frac{(Y_I + Y_{Xe})\alpha P - \left[\frac{DA V_G \lambda_I}{CA - Q_3 Q_4} + 0.9 Q_5 \right] (N_I)_G}{V_S \left[\lambda_{Xe} + \sigma_a(Xe)\phi \right] + \frac{DQ_4 L_1}{CA - Q_3 Q_4}} \quad (40)$$

$$\text{where } C = V_G \lambda_{Xe} + Q_3 + \frac{L_6}{\theta_L (K_{Xe})_L} \quad (41)$$

$$D = V_G \lambda_{Xe} + \frac{L_6}{\theta_L (K_{Xe})_L} \quad (42)$$

f_{Xe} calculated in all cases from $(N_{Xe})_S$ and Equation (4).

4.13 System 5-H (See Figure 5)

Removal of nuclear poison from main high pressure loop as I-135 dissolved in fuel solution; contacting of vapor with fuel solution plus passage of vapor bubbles directly from core to vapor space in pressurizer.

Equations (39), (40), (41), (42), and (43) apply.

Iodine balance around gas space through lines 4, 5, and 10.

$$Q_4 (N_I)_4 + Q_{10} (N_I)_{10} = (Q_5 + V_G \lambda_I) (N_I)_G \quad (43)$$

Assuming gas-liquid equilibrium in core

$$(N_I)_S = \theta_S (K_I)_S (N_I)_S \quad (44)$$

Balance around reactor through lines 1, 2, and 10

$$Y_I \alpha P = \left[V_S \lambda_I + L_1 + Q_{10} \theta_S (K_I)_S \right] (N_I)_S - \frac{L_2 (N_I)_4}{\theta_S (K_I)_S} \quad (45)$$

Iodine balance around mixer through lines 1, 2, 4, and 8 assuming compositions in 4 and 2 in equilibrium.

$$L_1 (N_I)_S + Q_8 (N_I)_8 = \left[\frac{L_2}{\theta_S (K_I)_S} + Q_4 \right] (N_I)_4 \quad (46)$$

Balance around condenser - assuming 90% absorption of iodine and neglecting radioactive decay:

$$Q_8(N_I)_8 = 0.1 Q_5 (N_I)_G \quad (47)$$

Combining Equations (43), (44), (45), (46), and (47)

$$(N_I)_G = \frac{Y_I \alpha P}{\left[\lambda_I V_S + L_1 + Q_{10} \theta_S (K_I)_S + \frac{L_2 Q_{10}}{Q_4} \right] \left[\frac{AB_2 - 0.1 Q_5}{L_1 + \frac{AQ_{10} \theta (K_I)_S}{Q_4}} \right] - \frac{L_2 B_2}{\theta_S (K_I)_S}} \quad (48)$$

Similarly, xenon balances around mixer, gas space, condenser, and Equation (29) results in

$$(N_{Xe})_S = \frac{(Y_I + Y_{Xe}) \alpha P - \left[\frac{DA V_G \lambda_I}{(L_2 A - Q_4 E)} + 0.9 Q_5 \right] (N_I)_G}{V_S \left[\lambda_{Xe} + \sigma_a (Xe) \phi \right] + \frac{DF}{AL_2 - Q_4 E}} \quad (49)$$

where

$$A = \text{Equation (38)}$$

$$B_2 = \frac{Q_5 + V_G \lambda_I}{Q_4} \quad (50)$$

$$C_2 = Q_5 + V_G \lambda_{Xe} \quad (51)$$

$$D = V_G \lambda_{Xe} + \frac{L_6}{\theta_L (K_{Xe})_L} \quad (52)$$

$$E = Q_5 - \frac{L_6}{\theta_L (K_{Xe})_L} \quad (53)$$

4.2 Calculations

4.21 Average Flux

(See Section 4.3 Nomenclature for many values used)

Assume fuel contains 4 g/l of U-233
0.4 g/l of U-235

$$\Sigma_f = N(25)\sigma(25) + N(23)\sigma(23)$$

$$= \left[\left(\frac{4}{233} \right) (346 \times 10^{-24}) + \frac{(0.4)(349 \times 10^{-24})}{235} \right] \frac{6.02 \times 10^{23}}{103}$$

$$= 3.94 \times 10^{-3} \text{ cm}^2/\text{ml}$$

$$\Phi_{\text{avg}} = \frac{P\alpha}{V_S \Sigma_f} = \frac{(50)(3.38 \times 10^{16})}{(4 \times 10^6)(3.94 \times 10^{-3})}$$

$$= 1.18 \times 10^{14} \text{ neutrons}/(\text{cm}^2)(\text{sec})$$

Equation for jet eductor performance from CF 56-7-12, p.9.

$$M\bar{V} (P_1 - P_2) = \frac{G}{E} (RT \ln P_4/P_3)$$

where:

M = gram moles liquid/min.

G = gram moles vapor/min.

\bar{V} = molal liquid volume

P_1 = entering liquid pressure

P_2 = discharge liquid pressure

P_3 = entering gas pressure

P_4 = discharge

E = efficiency of jet = 5% (assumed)

T = temperature = 280°C

$P_1 - P_2$ = 20 psi

$P_2 = P_4$ = 1500 psi

$P_4 - P_3$ = 1 psi

$$\frac{M}{G} = \frac{(\rho_S (1.8)(T)(10.73) \ln P_4/P_3)}{(0.32)(P_1 - P_2)(E)}$$

$$\begin{aligned}
 &= \frac{60.4(\rho_s)(T)\ln P_4/P_3}{E(P_1 - P_2)} \\
 &= \frac{(60.4)(0.85)(553)(\ln \frac{1500}{1499})}{(0.05)(20)} = 19.2
 \end{aligned}$$

4.23 Vapor Compressibility Factors

$$p V_G = n Z RT$$

$$n = \frac{V_G}{RT} \left[\frac{p_A}{Z_A} + \frac{p_B}{Z_B} + \frac{p_C}{Z_C} \right] = \frac{V_G}{RT} \frac{\pi}{Z_a}$$

where A = D₂O

B = D₂

c = O₂

a = average

Z = compressibility factor

Z_B = Z_C = 1 at reactor temperature and pressure

$$Z_a = \frac{\frac{p_A}{Z_A} + \frac{p_B}{Z_B} + \frac{p_C}{Z_C}}{\frac{\pi}{\pi + p_A \left(\frac{1}{Z_A} - 1 \right)}} = \frac{\pi}{\pi + p_A \left(\frac{1}{Z_A} - 1 \right)}$$

$$y_{D_2O} = \frac{p_{D_2O}}{Z_{D_2O} \left[\pi + p_{D_2O} \left(\frac{1}{Z_{D_2O}} - 1 \right) \right]}$$

$$y_{O_2} = \frac{p_{O_2}}{\pi + p_{D_2O} \left(\frac{1}{Z_{D_2O}} - 1 \right)}$$

$$y_{D_2} = \frac{p_{D_2}}{\pi + p_{D_2O} \left(\frac{1}{Z_{D_2O}} - 1 \right)}$$

$$K_{D_{20}} = \frac{H_{D_{20}}}{Z_{D_{20}} \left[\pi + P_{D_{20}} \left(\frac{1}{Z_{D_{20}}} - 1 \right) \right]}$$

$$K_{O_2} = \frac{H_{O_2}}{\pi + P_{D_{20}} \left(\frac{1}{Z_{D_{20}}} - 1 \right)}$$

$$K_{D_2} = \frac{H_{D_2}}{\pi + P_{D_{20}} \left(\frac{1}{Z_{D_{20}}} - 1 \right)}$$

Read values of $Z_{D_{20}}$ from Perry p. 353 (17) for proper values of T and $P_{D_{20}}$.

4.3 Nomenclature

Y_{Xe}	= xenon-135 fission yield, 0.059 atoms/fission
Y_I	= iodine-135 fission yield, 0.003 atoms/fission
λ_{Xe}	= xenon-135 decay constant, $2.09 \times 10^{-5} \text{ sec}^{-1}$
λ_I	= iodine-135 decay constant, $2.88 \times 10^{-5} \text{ sec}^{-1}$
N	= atomic concentration, atoms/cc
$\sigma_a(Xe)$	= absorption cross section, xenon-135, $= 3.1 \times 10^6$ barns
α	= 3.38×10^{16} fissions/sec/Mw
P	= Core power, 50 Mw
V_S	= volume of liquid in main core loop, 400 l (sp. power = 12.5 w/ml)
V_G	= volume of gas in main high pressure system
f_{Xe}	= xenon-135 poisoning ratio (see Equation (4))
ϕ	= 1.18×10^{14} neut/cm ² /sec average in 4000 l at 280°C
Π	= total pressure, psi
ρ	= density, gm/ml
R	= gas law constant
T	= absolute temperature
Z_a	= compressibility factor = $\frac{pv}{RT}$
L	= liquid flow rate, ml/sec
M	= liquid flow rate, g moles/sec
Q	= gas flow rate, ml/sec
G	= gas flow rate, g moles/sec
C	= condensate rate
MW	= atomic weight

$$\theta_L = \frac{(MW)_c}{\rho_c (Z_a)_G RT_G}$$

$$\theta = \frac{(MW)_S}{\rho_S (Z_a)_G RT_G}$$

H = Henry's Law constant, psi/mol fraction in liquid

K = Henry's Law constant, mol fraction in gas/mol fraction in liquid at $\pi = 1500$ psi, $T = 280^\circ\text{C}$

	<u>H</u>	<u>K</u>	
Oxygen in D_2O	2.65×10^5	140	(10)
Deuterium in D_2O	2.25×10^5	120	(10)
Oxygen in soup	2.76×10^5	146	(10)
Deuterium in soup	2.50×10^5	133	(10)
Iodine in soup	0.134×10^5	7	Reference (6)
Xenon in D_2O	1.4×10^5	73	(10)
Xenon in soup	1.4×10^5	73	(Assumed same as in D_2O)
Iodine in D_2O		0.2	(6)

Table I

Xenon Poisoning with Vapor Let-Down Systems
(Removal of Poison from Main Loop as Xenon-135)

System shown in Figure 3

	<u>D</u>		<u>E</u>	
L_1 , gpm liquid to contactor (jet) ^a	20	20	20	20
Q_3 , cfm gas to contactor ^a	2.1	2.1	2.1	2.1
Q_5 , cfm vapor ^a	0.022	0.21	4.17	4.17
L_6 , gpm condensate ^a	1.5	1.5	1.5	1.5
Q_7 , cfm vapor	-	-	0.95	0.0088
f_{Xe} , xenon-135 poison fraction	0.043	0.024	0.0089	0.041
$O_2 + D_2$ in vapor letdown, cfm(STP)	-	-	82	0.76

^a Volumes at 280°C and 1500 psi

^b Volume at 50°C and 1500 psi

Table II

Xenon Poisoning with Condensate Treatment
(Removal of Poison from Main Loop as Iodine-135)

System shown in Figure 5

	G		H		
L_1 , gpm liquid to contactor ^a	20	20	20	20	20
Q_3 , cfm gas to contactor	2.1	2.1	-	-	-
Q_8 , cfm gas to contactor	-	-	2.1	2.0	2.1
Q_{10} , cfm bubbles core to pressurizer	0	0	0	0.44	0.44
L_6 , gpm, condensate ^a	1.5	1.0	1.5	1.5	1.0
O_2 in vent gases, cfm(STP)	0.38	0.26	0.38	0.38	0.26
f_{Xe} , xenon-135 poison fraction	0.0105	0.015	0.011	0.010	0.011

^a Volumes at 280°C and 1500 psi

^b Assuming D_2 concentration in stream S is negligible

References

- (1) Belger, J. C. and Maak, R., ORNL CF-57-7-28.
- (2) Arthur D. Little, AECU-3349, "Gas Pressurization of HRT."
- (3) Rosenthal, M., and Fowler, T., ORNL CF-57-7-23.
- (4) Rosenthal, M., Personal Communication, August 23, 1957. (Report in Preparation).
- (5) Vitro Engineering Company
- (6) McNees, R., TID-7540, "HRP Civilian Power Conference, May 1-2, 1957," July 1957.
- (7) Stephan, E. F., et. al., BMI-705, "Explosion Limits of the H_2-O_2 -Water System at Elevated Temperatures, November 1, 1951.
- (8) Stephan, E. F., et al., BMI-1138, "Ignition Reactions in the H_2-O_2 -Water System at Elevated Temperatures, October 2, 1956.
- (9) Ordin, P. M., NACA-3935, "Hydrogen-Oxygen Explosion in Exhaust Ducting," April 1957.
- (10) Stephan, E. F., et al., BMI-1067, "The Solubility of Gases in Water and Aqueous Uranyl Salt Solutions at Elevated Temperatures and Pressures," Jan. 23, 1956.
- (11) Spiewak, I., Personal Communication, August 1957.
- (12) Haubenreich, P. N., CF 57-5-100, "Effects of Letdown Rates and O_2 Injection Rates on Xenon Poison Level and Excess Oxygen Concentration in the HRT," May 31, 1957.
- (13) Gift, E. H., "HRT Iodine Removal Bed," ORNL CF 57-9-50.
- (14) Ferguson, D. E., "Removal of Iodine from Homogeneous Reactors," ORNL CF 56-2-81, February 16, 1956.
- (15) Pigford, T. H., ORNL-1322, "Explosions and Detonation Properties of Mixtures of H_2 , O_2 and H_2O Vapors,"
- (16) Sherwood, T. K., and Pigford, R. L., "Absorption and Extraction", pp 144-148, 194-199, McGraw Hill Book Company, New York (1952).
- (17) Perry, J. H., ed., "Chemical Engineers' Handbook," 3rd ed., McGraw Hill Book Company, New York (1950).

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