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ENGINEERING DEVELOPMENT OF
AN ABSORPTION PROCESS FOR THE CONCENTRATION
AND COLLECTION OF KRYPTON AND XENON
FOURTH SUMMARY PROGRESS REPORT
JULY THROUGH DECEMBER, 1968

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UNION CARBIDE CORPORATION
NUCLEAR DIVISION
OAK RIDGE GASEOUS DIFFUSION PLANT

operated for the **ATOMIC ENERGY COMMISSION** *under* **U. S. GOVERNMENT** **Contract W-7405 eng 26**

**UNION
CARBIDE**

OAK RIDGE GASEOUS DIFFUSION PLANT
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124

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A B S T R A C T

A continuous selective absorption process for the removal of krypton and xenon from contaminated gas streams by preferential dissolution in a fluorocarbon solvent is being developed at the Oak Ridge Gaseous Diffusion Plant. The most important part of the ORGDP program is pilot plant testing, which is being conducted to determine feasibility and to define the main performance parameters in terms of the operating variables. The pilot plant work is supplemented by conceptual plant design work and by process optimization studies.

During this report period, in which refrigerant-12 was used as the process solvent, pilot plant shakedown was completed, and the first phase of the formal testing program was begun. The conceptual design work was limited to further investigation of solvent radiolytic decomposition in the absorption process. The optimization studies included analyses of three absorption plants designed to treat off-gases from fuel reprocessing systems.

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TABLE OF CONTENTS

	<u>PAGE</u>
INTRODUCTION	9
SUMMARY	9
PROJECT DEFINITION	10
PILOT PLANT STUDIES	10
Flow Sheet Development	10
Equipment Design	10
Operation	11
CONCEPTUAL DESIGN STUDIES	13
General Procedure for Estimating Solvent Degradation Rate	13
Simplified Procedures for Estimating Solvent Degradation Rate. .	16
Long Cooling Time Case	18
Short Cooling Time Case	19
OPTIMIZATION STUDIES	21
STATUS AND FUTURE PLANS	21
BIBLIOGRAPHY	24

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LIST OF TABLES

<u>Table</u>	<u>Page</u>
I Absorption System Performance Data	12
II Summary of G-Values for Solvent Degradation	14
III Decay Data for Krypton-85 and Xenon-133	17
IV Estimated Degradation Rates for Long Cooling Time Processing Situations	19
V Estimated Degradation Rates for Short Cooling Time Processing Situations	20
VI Optimization Case Study Feed Gas Flows and Compositions	22
VII Column Operating Temperatures and Pressures for Case Studies	22
VIII Case Study Results	23

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ENGINEERING DEVELOPMENT OF AN ABSORPTION PROCESS
FOR THE CONCENTRATION AND COLLECTION OF KRYPTON AND XENON
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JULY THROUGH DECEMBER, 1968

The Oak Ridge Gaseous Diffusion Plant is developing a process for the continuous removal of fission product krypton and xenon from contaminated gas streams. Such treatment, which eliminates the need to discharge these radioactive isotopes directly to the atmosphere, will probably be required for environmental protection in the future in a number of nuclear processing situations.

The process being studied at the ORGDP is based on selective absorption of krypton and xenon in liquid fluorocarbon solvents, such as refrigerant-12. Incoming, contaminated gas requiring treatment is fed to a packed absorption column, where it is contacted with downflowing solvent. Since krypton and xenon are preferentially dissolved in the liquid, the off-gas from the absorber can be safely vented. The loaded solvent flowing from the absorber is then fractionated in auxiliary equipment to yield, ultimately, a low flow rate product gas stream, rich in krypton and xenon, and a lean solvent stream for recycle to the absorber column.

The main part of the ORGDP program is demonstration of the process via pilot plant studies aimed at determining feasibility and at defining the relationships between performance parameters and operating variables. The pilot plant effort is supplemented by conceptual plant design work and by process optimization studies. Progress made in each of these areas during the first half of FY-1969 is summarized in the following sections. Previous work on this project is described in three earlier progress reports^[1,2,3] and in a recent paper^[4].

SUMMARY

During this report period, the major emphasis was placed on completing the pilot plant shakedown work and on initiating the first phase of the formal testing program. The shakedown was successfully completed during the first three months of this period, after which an on-line gas chromatograph, for use in determining compositions of the various process gas streams, was installed and calibrated. Then, refrigerant-12, the solvent being used in the first series of tests, was charged to the system, and the formal experimental work was begun. To date, seven runs have been made. In these runs, the absorber column was operated at a pressure of 416 psia and at temperatures between minus 32 and minus 24°F. Solvent flow rates of 0.75 and 1.0 gpm were used, while the absorber feed gas flow rate was varied between 9.5 and 20.6 scfm. Krypton removals between 75.02 and 99.81% were observed, corresponding to absorber column krypton decontamination factors between about 4 and 525. From an operating standpoint, system performance has also been very good.

The conceptual design work was limited to a continuation of studies, begun during the previous report period, of the amount of solvent decomposition which would occur in the absorption process because of exposure of the liquid to radioactive gases. Even though very conservative assumptions were made, the calculations still indicated that, for the applications of the process now envisioned, solvent degradation will probably not be a significant problem.

In the optimization study portion of the program, the gas cleaning requirements of a fuel reprocessing plant were considered. Three possible reprocessing plant off-gas flow rates were looked at; namely, 20, 100, and 500 scfm. In each case, conditions for and capital costs of absorption plants designed for about 99% recovery of the noble gases were determined. These costs were near \$100,000; \$260,000; and \$830,000 for the three cases, increasing, of course, with the flow rate.

PROJECT DEFINITION

There were no changes in basic project orientation during this report period. Both the experimental and study efforts have been planned so that the results will be applicable to a variety of krypton-xenon removal problems.

PILOT PLANT STUDIES

M. J. Stephenson, L. W. Anderson, D. L. Burkett

As described in the previous progress reports, the ORGDP noble gas removal pilot plant consists of three columns--an absorber, a fractionator, and a stripper--plus attendant heat transfer and pumping equipment. The absorber and fractionator columns are 3 inches in diameter and are packed to a height of 9 feet; the stripper column is 6 inches in diameter and contains 8 feet of packing. The nominal capacity of the pilot plant is 15 scfm of gas feed to the absorber, the lower temperature limit is minus 100°F, and the highest pressure attainable is 600 psi. Complete flow sheet descriptions and design details are presented in the earlier progress reports [1,2,3].

Flow Sheet Development

No basic flow sheet changes were made during this report period.

Equipment Design

One of the encouraging aspects of the shakedown work was that no equipment design changes were indicated.

Operation

Shakedown tests, using refrigerant-11 as the solvent, were successfully completed during the first three months of this report period. From a hydraulics standpoint, the system was generally found to perform satisfactorily, with only minor valving changes indicated. Some instrumentation problems mentioned in the previous report[3] were also corrected.

Additionally, considering the observed separation data, refrigerant-11 continues to appear promising as a higher temperature substitute for the refrigerant-12 process solvent[3]; consequently, it is planned to study the performance of this solvent further following the refrigerant-12 tests.

After the shakedown tests, the pilot plant system was cleaned, vacuum-dried, and charged with refrigerant-12. Then, while the chromatograph was being installed and calibrated, some scoping tests with this solvent were made. It was quickly found that under some conditions originally included in the test plan, i.e., at relatively low temperatures, high pressures, and high absorber liquid-to-gas flow rate ratios, krypton removals were even more complete than first expected. For example, with krypton concentrations in the absorber feed gas of 1000 ppm or more, the krypton content of the vent stream was in some cases below detectable limits (less than 5 ppm). While such complete removals (greater than 99.99%) are quite impressive, they were not desired in the pilot plant testing program. In the pilot plant work, it is necessary to have accurately detectable quantities of krypton in the absorber outlet gas stream; otherwise, the mass transfer calculations required for process analysis and the correlation of performance indicators are not possible. Consequently, the formal run plan was revised so that conditions of such complete recovery would be avoided. This revision was also supported by the results of the process optimization studies, which indicated that absorption temperatures considerably above the originally planned minus 94°F are also favored for economic reasons.

Next, the refrigerant-12 testing program was initiated. To date, seven runs have been completed, and the results are summarized in table I. The purpose of this particular set of tests was to provide an initial indication of how the absorber column performance varies with gas and liquid flows for approximately constant operating pressure and temperature.

The first five tests were conducted with gas flow rates near 10, 15, and 20 scfm; varying krypton concentration; and a constant solvent flow of 0.75 gpm. For the sixth and seventh runs, the gas flow was held near 15 scfm, the liquid rate was increased to 1.0 gpm, and the krypton concentration was maintained at a relatively constant level. The absorber column was, in each case, operated near 416 psia and in the temperature range from minus 32 to minus 24°F. Also, for each of the runs, the fractionator was operated at about 33°F and 45 psia, and the stripper at approximately 2°F and 25 psia.

TABLE I
ABSORPTION SYSTEM PERFORMANCE DATA

	Run Number						
	1	2	3	4	5	6	7
Average Absorber Temperature, °F	-24	-25	-30	-32	-31	-26	-26
Absorber Pressure, psia	416	416	416	416	416	416	416
Solvent Flow Rate, gpm	0.75	0.75	0.75	0.75	0.75	1.0	1.0
Absorber Feed Gas:							
Flow Rate, scfm	9.5	9.6	15.1	15.0	20.6	15.2	15.5
Krypton Concentration, ppm.	990	1670	125	305	60	1130	1070
Absorber Off-Gas:							
Flow Rate, scfm	5.5	6.1	9.4	8.5	14.7	9.7	9.2
Krypton Concentration, ppm	5	5	5	8	21	23	15
Average L/G Ratio, mole basis	4.52	4.25	2.81	3.04	1.92	3.58	3.70
Krypton Removal in Absorber, %	99.71	99.81	97.51	98.51	75.02	98.70	99.17
Absorber Column Krypton Decontamination Factor	345	526	40	67	4	77	120

The results obtained to date indicate that, for a given solvent flow, the amount of krypton removed in the absorber column is a strong function of the liquid-to-gas flow rate ratio. As expected, increasing the relative proportion of solvent fed to the column generally increases the overall krypton removal efficiency. One of the minor disadvantages of operating with a higher solvent flow, however, is the fact that increased amounts of oxygen, nitrogen, and argon are carried over with the noble gases to the fractionator, thereby increasing the load on that unit.

A very encouraging aspect of the testing program, in addition to the krypton removal data, is that operation of the pilot plant has continued to be relatively troublefree. Including the shakedown work, about 2,000 on-stream hours have been logged, with no incidents forcing prolonged shutdowns.

CONCEPTUAL DESIGN STUDIES

K. E. Habiger, B. F. Crump

The extent of solvent degradation due to exposure to radioactive nuclides is always an important consideration in radiochemical process design. Accordingly, during this report period, calculations aimed at estimating the magnitude of this problem for the krypton-xenon absorption process were continued. In the previous report^[3], results of a literature survey for data concerning the radiation stability of refrigerant-11 and -12 were presented; these data are also summarized in table II. In this report, these G-values are used to illustrate procedures for predicting solvent degradation rates for possible plant cases.

General Procedure for Estimating Solvent Degradation Rate

In a particular equipment item, the rate at which the solvent inventory will undergo radiolytic decomposition is:

$$D_P = \frac{(5.308)(10^{-3})(G_P)}{x_s} \left\{ \sum_j [x_j (\sum_i f_{ji} A_{ji} E_{ji} M_{ji})] \right\}, \quad (1)$$

where D_P = solvent degradation rate applicable to inventory in vessel P, percent of inventory decomposed per day;

G_P = G-value appropriate for vessel P's operating conditions, molecules solvent decomposed per 100 ev of absorbed energy;

x_s = solvent mole fraction in liquid phase;

x_j = average mole fraction of gaseous element j dissolved in liquid;

f_{ji} = fraction of element j which is radioactive isotope i;

TABLE II

SUMMARY OF G-VALUES FOR SOLVENT DEGRADATION*

Temperature, °F	G-Value, molecules solvent destroyed per 100 ev of absorbed energy**	
	<u>Refrigerant-12</u>	<u>Refrigerant-11</u>
- 94		0.78
32	3.58	3.82
95 to 100	3.6	4.6

The refrigerant-11 and -12 data, if taken together, can be approximated by the equation:

$$G = 120 e^{-\frac{3594}{RT}},$$

where T is in °R, and R = 1.987 Btu/lb·mole-°R.

* Merriman, J. R., and Pashley, J. H., *Engineering Development of an Absorption Process for the Concentration and Collection of Krypton and Xenon, Third Summary Progress Report, January through June, 1968*, Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, March 27, 1969 (K-1770).

** It is assumed in this table that one solvent molecule is destroyed for each halogen ion produced. This is a somewhat conservative assumption since it does not account for the possibility that more than one halogen ion may be removed from a single solvent molecule.

A_{ji} = activity of species ji , curies per gram;

E_{ji} = effective energy release from species ji , Mev per disintegration; and

M_{ji} = molecular weight of isotope ji .

The average rate of solvent degradation for the entire plant, then, is attained by applying equation (1) to all equipment items used in the plant.

$$D_{TOT} = \frac{\sum_P D_P I_P}{\sum_P I_P}, \quad (2)$$

where D_{TOT} = overall average solvent degradation rate, percent per day, and

I_P = inventory of vessel P , any units.

Thus, for a given plant case, a vessel-by-vessel accounting is made of the extent of solvent decomposition, as follows:

1. Based on the operating temperature for the unit being considered, an appropriate G-value is obtained, considering the data presented in the previous report[3] and summarized in table II.
2. From the operating conditions for the vessel and material balances, values of x_i for those gaseous elements which have radioactive isotopes are computed.
3. The nature of the gas cleaning job being accomplished determines which radioactive isotopes are present in significant quantities and their relative amounts. Accordingly, f_{ji} for all isotopes important for the particular case can be obtained.
4. Values of A_{ji} and E_{ji} for the isotopes being considered are readily available from any of several data compilations.
5. The operating solvent inventory of a particular equipment item can be computed based on sizing parameters.
6. Equation (1) can then be used systematically for each vessel, and its associated piping and hold volume, to provide an estimate of the solvent decomposition rate for each sector of the process. Finally, equation (2) provides the overall plant average solvent degradation rate.

As an absorption plant is being designed for some specific krypton-xenon removal situation, the procedure just outlined should be used to estimate the solvent degradation rate for that particular application of the

process. In general, however, it is also possible to provide some upper limit estimates of the extent of solvent destruction without reference to any specific plant design. These estimates, described in the following sections, are made possible through the use of certain simplifying, yet still conservative, assumptions.

Simplified Procedures for Estimating Solvent Degradation Rate

Two separate cases can be looked at as typifying *best* and *worst* processing situations from the standpoint of solvent radiolytic decomposition. In the first of these, which might be represented by a light-water reactor fuel reprocessing plant off-gas treatment system, cooling times are large enough so that essentially all of the radioactive isotopes except krypton-85 have decayed away. The second case, on the other hand, might represent a situation where the absorption system feed gas is taken directly from a nuclear reactor blanket containing equilibrium production concentrations of several active krypton and xenon isotopes. Thus, in this case, other isotopes, primarily xenon-133, must also be considered. To assist in the following discussion of these cases, decay data for krypton-85 and xenon-133 are listed in table III.

Now, some conservative assumptions can be made, as follows:

1. Since the degradation rate increases directly with the G-value, and since the G-value increases with increasing temperature, the number corresponding to the highest temperature occurring in the process can be assumed to apply to the entire plant inventory. (Because of the large differences between temperatures of the hottest and coldest portions of the plant, this assumption should be very conservative.) With refrigerant-12 as the solvent, the highest temperature, about 32°F, is the fractionator temperature; the reported G-value for this material at this temperature is 3.58.
2. From equation (1), the decomposition rate is also seen to increase directly with increasing average dissolved gas mole fraction, x_j . In turn, for each gas, this number increases with increasing pressure, increasing gas-phase mole fraction, and decreasing temperature. In the absorption process, the highest pressure, the largest mole fraction of radioactive isotopes in the gas phase, and the lowest temperatures all occur at the inlet of the absorber column*. Furthermore, the average liquid phase dissolved gas mole fractions in the

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* This is true in all cases except for small portions of the fractionator and stripper final condensers which are about as cold as the absorber. In these units, the active gas mole fractions may considerably exceed the absorber column values at some point near the exits; however, the pressures are lower, and more important, the liquid solvent inventory at these points is essentially zero.

TABLE III

DECAY DATA FOR KRYPTON-85 AND XENON-133

	<u>Krypton-85</u>	<u>Xenon-133</u>	<u>Reference or Comments</u>
Half-Life	10.60 years	5.27 days	*
A_{ij} , Specific Activity, curies/gram	397	186,000	*
Gamma Releases, Mev/dis	0.41% @ 0.514	Taken as 99.3% @ 0.081	**
Maximum Beta Release, Mev/dis	0.67	0.346	**
E_{ij} , Total Energy Released to Solvent, Mev/dis	0.225	0.196	Average beta taken as 1/3 of endpoint, or maximum, value
* Goldstein, G., and Reynolds, S. A., "Specific Activities and Half-Lives of Common Radionuclides", <i>Nuclear Data</i>, A, 1, 5, p. 435-452 (July, 1966). ** Lederer, C. M., <u>et al.</u>, <i>Table of Isotopes</i>, Sixth Edition, John Wiley and Sons, New York, First Corrected Printing (March, 1968).			

absorber can never exceed the values which would correspond to equilibrium with this feed gas. Accordingly, another conservative assumption is to take, as applicable to all of the process inventory, dissolved gas mole fractions in equilibrium with the absorber feed gas:

$$x_j = \frac{(y_j) \text{ absorber feed}}{(k_j) \text{ absorber column}}, \quad (3)$$

where y_j = mole fraction of element j in the absorber column feed gas*, and

k_j = equilibrium constant for element j at the absorber conditions.

When it is realized that over one-half of the absorption process inventory will probably be stripped solvent, held in the solvent reservoirs and containing at most only trace amounts of krypton and xenon, this assumption is, in particular, seen to be very conservative.

3. Additionally, it can be assumed that all of the gamma-plus-beta radiation emitted by the decaying isotopes is absorbed by the solvent.
4. As was noted in table II, it can be further assumed that one solvent molecule is destroyed for each halogen ion produced by the irradiation. That is, the possibility that more than one halogen ion might be removed from the same solvent molecule is ignored.

Using these assumptions of maximum G-value, maximum radioactive gas content, and maximum energy absorption, the two cases previously mentioned can be studied further.

LONG COOLING TIME CASE

For light-water reactor fuel, krypton-85 typically comprises about 7% of the total krypton produced during irradiation[5], so that f_{j1} for this isotope is nearly equal to 0.07 in many cases. Therefore, the degradation rate under the assumptions noted above reduces to

$$D_{TOT} \doteq 10.2 x_1 \doteq 10.2 \frac{y_1}{k_1}. \quad (4)$$

Also, for krypton dissolved in refrigerant-12, the equilibrium constant, k_1 , is 2.32 at 14 atmospheres and minus 94°F and about 2.7 at 30 atmospheres and minus 25°F[6]; therefore, a value of 2.5 should be

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* The absorber feed is made up of fresh feed entering the process from some nuclear facility plus recycle gas from the fractionator.

representative of absorber conditions of most interest in refrigerant-12 based processes. Using this value, degradation rates are listed in table IV for various concentrations of krypton in the absorber feed gas stream. It can be seen that, for the concentrations most likely to be encountered in actual processing situations, the solvent degradation rate is negligible, less than 0.02% per day.

TABLE IV
ESTIMATED DEGRADATION RATES
FOR LONG COOLING TIME PROCESSING SITUATIONS

Krypton Concentration in Absorber Feed Gas, y_i , ppm	Estimated Solvent Degradation Rate, percent per day
50	0.0002
100	0.0004
500	0.002
1000	0.004
5000	0.02

SHORT COOLING TIME CASE

Prediction of degradation rates for this case is more difficult than for the long cooling time situation. This is due primarily to the fact that, for accurate calculations, several krypton and xenon isotopes need to be considered. To compute the concentrations of these isotopes entering the absorption system, not only must the irradiation conditions for the associated reactor be used to obtain equilibrium production quantities, but also the specific transport mechanisms and hold times associated with movement of each isotope first from its formation site in the fuel to the containment gas and then to the absorption process must be used to determine the extent of decay and hence to provide a steady-state profile of the absorber feed*. Even then, in-process decay should also be accounted for in the case of some short half-life species. Based on some preliminary

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* For example, in a molten salt reactor system, fission product gases are formed in the salt and then diffuse, or are carried by bubbles, etc., to the blanket gas. This requires some finite time, during which decay occurs.

estimates for a few irradiation schemes, however, it appears that between 0.2 and 1% of the xenon will be radioactive and that the primary xenon isotope will be xenon-133. Thus, for these situations, f_{ji} for this isotope will be between 0.002 and 0.01. Following a development similar to that for the previous case, considering both krypton-85 and xenon-133 contributions*, and using a xenon equilibrium solubility constant of 0.7, degradation rates for both values of f_{ji} were computed. Presented in table V, these rates do not appear to be too significant except when a relatively large fraction of the xenon processed is radioactive and when the xenon content of the absorption system feed gas is appreciable. In such instances, the rigorous procedure discussed earlier should be used to predict a more accurate value without making the many conservative assumptions made here, in order to determine whether in-line solvent purification equipment is necessary or if other adjustments are required.

TABLE V
ESTIMATED DEGRADATION RATES
FOR SHORT COOLING TIME PROCESSING SITUATIONS

Xenon Concentration in Absorber Feed Gas*, y_2 , ppm	Estimated Solvent Degradation Rate, percent per day	
	$f_{21} = 0.002$	$f_{21} = 0.01$
50	0.015	0.068
100	0.028	0.13
500	0.13	0.66
1000	0.26	1.3
5000	1.3	6.6

* The krypton concentration was taken at 500 ppm, and the solvent degradation rate corresponding to this value was added to the xenon contribution.

* Actually, in some cases krypton-85 may not be the most significant krypton isotope. However, it appears that, in general, the xenon-133 contribution to degradation is the most important one by a wide margin.

In summary, even when using some very conservative assumptions to estimate solvent radiolytic decomposition rates, solvent degradation does not appear to be a significant problem in most of the cases felt to represent future absorption process applications. It should be noted again, however, that it would certainly be appropriate to include an item-by-item estimate of the degradation rate for each specific plant case as a part of the plant design effort, to be sure that there are no unusual problems.

OPTIMIZATION STUDIES

S. Blumkin, E. Von Halle, D. J. Roberts

The computer program developed for modeling the operation of a three-column gas absorption plant and computing the associated capital costs was revised somewhat to facilitate parametric case studies. Given the operating conditions (pressure, column temperature, partial and total condenser temperatures, number of stages per column, and reboil rate) of the absorber, fractionator, and stripper systems, plus initial guesses for the flow rates and compositions of two internal streams, the flow rates and compositions of all plant streams are computed and balanced by iterative techniques for up to a five-component gas feed. These results are then used to compute plant capital cost.

Recently, three cases have been investigated. These cases involved the removal of xenon and krypton from air in a reactor fuel reprocessing plant situation. Three feed flow rates, 20, 100, and 500 scfm, were investigated, corresponding to three diluent air throughputs for the same noble gas release rate. A noble gas recovery of about 99% was desired. The input parameters are listed in tables VI and VII, and the results are summarized in table VIII. The estimated capital costs were about \$100,000 for the 20 scfm case; \$260,000 for the 100 scfm feed rate; and \$830,000 for the 500 scfm plant.

Originally, the absorber temperature and pressure were assumed to be minus 94°F and 206 psia. In attempts to arrive at lower plant costs, it was found that the large refrigeration costs necessitated increasing the absorber temperature to a value of minus 4°F, but a higher pressure of 588 psia was then needed to obtain the desired high recovery. The resultant refrigeration cost savings, however, far outweighed the increased compressor costs. The initial estimate of ten stages per column was increased to twelve or sixteen for high recovery with negligible incremental cost increases.

STATUS AND FUTURE PLANS

The first phase of the pilot plant testing program will be continued; it is expected that this part of the project can be completed during the

TABLE VI
OPTIMIZATION CASE STUDY
FEED GAS FLOWS AND COMPOSITIONS

<u>Case</u>	<u>Flow Rate, scfm</u>	<u>Feed Gas Compositions, mole %*</u>	
		<u>Xenon</u>	<u>Krypton</u>
1	20	0.45	0.05
2	100	0.09	0.01
3	500	0.018	0.002

* The remainder was assumed to be air.

TABLE VII
COLUMN OPERATING TEMPERATURES AND PRESSURES
FOR CASE STUDIES*

<u>Column</u>	<u>Temperature, °F</u>	<u>Pressure, psia</u>
Absorber	- 4	588
Fractionator	32	44
Stripper	12	29

* For all cases.

TABLE VIII
CASE STUDY RESULTS

	Case Number		
	1	2	3
Product Rate, scfm	0.321	0.318	0.318
Number of Stages*	16	12	16
Product Composition, mole %			
Xenon	28.04	28.27	28.33
Krypton	3.09	3.05	3.07
Noble Gas Recovery, %			
Xenon	99.99	99.95	99.99
Krypton	99.05	97.04	99.99
Estimated Capital Cost (1000's \$)	103.6	258.2	831.7
* For each column.			

next report period. The conceptual design effort will be suspended temporarily, until further guidelines are established. The optimization work will be continued with more case studies.

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