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ENGINEERING DEVELOPMENT OF AN ABSORPTION PROCESS FOR THE CONCENTRATION AND COLLECTION OF KRYPTON AND XENON FIFTH SUMMARY PROGRESS REPORT JANUARY THROUGH JUNE, 1969

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UNION CARBIDE CORPORATION
NUCLEAR DIVISION
OAK RIDGE GASEOUS DIFFUSION PLANT

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ENGINEERING DEVELOPMENT OF AN ABSORPTION PROCESS FOR THE CONCENTRATION AND COLLECTION OF KRYPTON AND XENON

FIFTH SUMMARY PROGRESS REPORT
JANUARY THROUGH JUNE, 1969

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A B S T R A C T

A selective process for removing 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 technique being studied is a continuous absorption-stripping process. Pilot plant demonstration of this process forms the major part of the ORGDP program, although conceptual plant design work and process optimization studies supplement the experimental effort.

During this report period, the first phase of the testing program, in which refrigerant-12 was used as the process solvent, was successfully completed. Pending further definition of process application guidelines, the conceptual plant design part of the project was temporarily suspended. The optimization studies were continued, however, with the investigation of an absorption plant suitable for removing the krypton and xenon which might be present in the argon cover gas of a vented fuel breeder reactor.

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ENGINEERING DEVELOPMENT OF AN ABSORPTION PROCESS FOR THE CONCENTRATION AND COLLECTION OF KRYPTON AND XENON FIFTH SUMMARY PROGRESS REPORT JANUARY THROUGH JUNE, 1969

The Oak Ridge Gaseous Diffusion Plant is engaged in development work aimed at demonstrating a process for the removal of krypton and xenon from contaminated gas sources. The need for this process has resulted because the safe handling of gas streams containing radioactive krypton and xenon isotopes is one of the important problems which will be faced by the nuclear industry in the near future. This disposal problem will arise in a number of instances, including large-scale nuclear fuel reprocessing and cleanup of reactor core cover gases.

The gas cleaning technique being developed at the ORGDP is a version of a conventional gas-liquid absorption-stripping process. Since krypton and xenon are appreciably more soluble than other air constituents in some fluorocarbon solvents, such as refrigerant-12, these two gases can be preferentially removed from contaminated streams by selective absorption. This provides an off-gas stream, substantially free of krypton and xenon, which can be safely vented, and a loaded solvent stream in which the noble gases are dissolved. This solvent stream, in turn, can subsequently be dealt with in one or more stripping operations to yield (a) a low flow rate gas product stream which is rich in krypton and xenon and is suitable for storage or perhaps further processing; and (b) an essentially pure solvent stream, for recycle to the absorber.

The main part of the ORGDP noble gas removal project is the engineering-scale investigation of the continuous absorption process in a pilot plant facility. This work is supplemented by conceptual plant design efforts and by process optimization studies. The overall goals are to determine the feasibility and reliability of the process, to establish how the separation performance varies with the important operating parameters, and to define economic operating ranges for different plant applications of the process.

Progress made in the main program areas during the second half of FY-1969 is summarized in the following sections. For discussions of previous work on this project, reference can be made to earlier progress reports[1,2,3,4] and to a summary paper[5].

SUMMARY

During this report period, the first phase of the pilot plant experimental work was completed successfully. With refrigerant-12 as the process solvent, thirty-four tests (including seven during the prior report period) were made under different sets of operating conditions. The absorption temperature was varied between minus 77 and minus 21°F, and the absorption pressure ranged from 164 to 437 psia, while solvent flow rates of

0.75, 1.0, and 1.25 gpm were used. The gas feed rate to the absorber column varied from 9.5 to 22.3 scfm, and the krypton concentration in this stream ranged from 42 to 8800 ppm. In the tests, krypton removals between 71.52 and 99.90% were observed, corresponding to absorber column krypton decontamination factors between about 3.5 and 1000. In addition to yielding good separation data, the pilot plant operations continued to be very smooth.

Since firm guidelines have still not been established for plant applications of this process, the conceptual design portion of the program was suspended during this report period. The optimization studies were continued, however, with the investigation of an absorption plant capable of removing krypton and xenon from an argon blanket gas which, for example, might be used with some types of nuclear reactors. The feed rate to the absorption process was assumed to be 10 scfm argon, and three product stream flow rates (0.02, 0.10, and 1.0 scfm) were then looked at for this plant. In each of the three cases, the estimated plant capital cost was slightly less than $100,000.

PROJECT DEFINITION

There were no changes in project orientation during this report period.

PILOT PLANT STUDIES

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

The ORGDP noble gas removal pilot plant utilizes three packed columns, plus associated heat transfer and pumping equipment. The absorber column, which is 3 inches in diameter and contains 9 feet of packing, is sized to accommodate a nominal gas feed flow of 15 scfm and to operate at temperatures as low as minus 100°F and pressures as high as 600 psia. The fractionator column is the same size as the absorber column, but this unit is designed to operate at higher temperatures and lower pressures. The stripper has a diameter of 6 inches and a packed height of 8 feet and is also sized for higher operating temperatures and lower operating pressures than the absorber. Complete flow sheet and equipment design details have been discussed previously[1,2,3].

Flow Sheet Development; Equipment Design; and Construction, Procurement, and Installation

No work in any of the above areas was required during this report period.

Operation

The first phase of the formal pilot plant experimental program was finished during this report period, with the successful completion of a series of thirty-four tests in which refrigerant-12 was used as the process solvent. The test conditions and results are summarized in table I. As indicated, the absorber column was operated at temperatures between minus 77 and minus 21°F, at pressures between 164 and 437 psia, and with solvent flow rates ranging from 0.75 to 1.25 gpm. The flow rate of the absorber column feed gas varied from 9.5 to 22.3 scfm, with krypton concentrations in this stream between 42 and 8800 ppm. Krypton removals between 71.52 and 99.90% were achieved, corresponding to absorber column krypton decontamination factors in the range of 3.51 to 1000. Even more complete krypton removals have been attained, but these tests were not included in the formal run format, for reasons noted in the previous progress report[4].

TABLE I

SUMMARY OF REFRIGERANT-12 TEST PARAMETERS

Variable	Test Range
Absorber Temperature, °F	- 77 to - 21
Absorber Pressure, psia	164 to 437
Solvent Feed Rate, gpm	0.75 to 1.25
Absorber Column Feed Gas	
Flow Rate, scfm	9.5 to 22.3
Krypton Concentration, ppm	42 to 8800
Average Absorber Column L/G Ratio, mole basis	1.72 to 5.93
Krypton Removal, %	71.52 to 99.90
Absorber Column Krypton Decontamination Factor	3.51 to 1000

In these tests, the fractionator and stripper systems were operated to provide a krypton feed stream for the absorber column. The operating temperatures of these units were near 33°F for the fractionator and 2°F for the stripper, while the pressures were about 45 and 25 psia, respectively.

Detailed data for each of the refrigerant-12 tests are listed in table II. These data indicate that the amount of solvent fed to the absorber, relative to the gas feed rate, is one of the key factors influencing krypton removal. Increasing the L/G ratio generally increased the krypton removal percentage at the various temperature and pressure levels. Both the absorption temperature and the absorption pressure were also seen to affect the krypton removal; krypton absorption was generally higher at lower temperatures and at higher pressures. In addition, for the same liquid and gas flow rates, selectivity for krypton appeared to be improved at the lower temperatures, for a given pressure; on the other hand, at higher pressures and for a given temperature, increased amounts of oxygen, argon, and nitrogen were absorbed with the krypton, thereby increasing the load on the fractionator system.

The first phase of the testing program is considered to be very successful, not only because it has been shown that the krypton removals required for essentially all plant applications of the process can be readily achieved, but also because the pilot plant has continued to operate well, with a minimum of forced downtime. Including the shakedown test, the pilot plant has now been operated for about 4500 hours. The data from the refrigerant-12 testing program are currently being analyzed further, and a topical report covering this phase of the work is being prepared.

CONCEPTUAL DESIGN STUDIES

K. E. Habiger, B. F. Crump

Work in this area was suspended during this report period, pending further definition of specific plant cases.

OPTIMIZATION STUDIES

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

Preliminary case studies aimed at estimating costs for absorption plants designed for removal of krypton and xenon from an argon stream were completed during this report period. The input data for these calculations, which might typify gas cleaning requirements for a vented fuel LMFBR with an argon blanket, are listed in table III. As can be seen, the basic separation job was reduction in activity of a 10 scfm argon stream from 567 to 0.03 curies/scf.

These cases were looked at, corresponding to three different concentrated product stream flow rates. The plant design and operating parameters used in these three sets of calculations are given in table IV. Each plant outlined is capable of achieving at least the target degree of decontamination, but the product composition varies considerably, as expected, with product flow rate, with the higher flow cases progressively more dilute in krypton and xenon. This is because the product krypton and xenon

TABLE II

REFRIGERANT-12 TEST DATA

Run Number	Average Absorber Conditions: Temperature, °F	Average Absorber Conditions: Pressure, psia	Solvent Flow Rate, gpm	Absorber Feed Gas: Flow Rate, scfm	Absorber Feed Gas: Krypton Concentration, ppm	Absorber Off-Gas: Flow Rate, scfm	Absorber Off-Gas: Krypton Concentration, ppm	Average L/G Ratio, mole basis	Krypton Removal in Absorber, %	Absorber Column Krypton Decontamination Factor
1	- 24	416	0.75	9.5	990	5.5	5	4.52	99.71	345
2	- 25	416	0.75	9.6	1670	6.1	5	4.25	99.81	526
3	- 30	416	0.75	15.1	125	9.4	5	2.81	97.51	40.2
4	- 32	416	0.75	15.0	305	8.5	8	3.04	98.51	67.1
5	- 31	416	0.75	20.6	60	14.7	21	1.92	75.02	4.00
6	- 26	416	1.0	15.2	1130	9.7	23	3.58	98.70	76.9
7	- 26	416	1.0	15.5	1070	9.2	15	3.70	99.17	120
8	- 24	437	1.0	15.0	3080	6.5	19	4.71	99.73	370
9	- 23	434	1.0	15.4	1640	7.1	8	4.43	99.78	455
10	- 23	338	1.0	14.9	8800	9.9	470	3.55	96.45	28.2
11	- 22	338	1.0	14.9	7700	9.7	400	3.59	96.62	29.6
12	- 25	338	1.25	15.0	690	9.8	16	4.42	98.48	65.8
13	- 26	260	1.0	14.4	8780	10.9	1360	3.38	88.28	8.53
14	- 26	260	1.0	14.9	8200	11.5	1680	3.23	84.19	6.33
15	- 26	260	1.0	14.6	5820	12.0	715	3.15	89.90	9.90
16	- 43	260	1.0	15.7	1620	12.0	272	3.14	87.17	7.79
17	- 45	260	1.0	15.5	6900	12.2	1260	3.13	85.63	6.96
18	- 41	260	1.0	15.8	5800	12.6	1090	3.02	85.01	6.67
19	- 53	260	1.25	15.3	1320	11.0	22	4.18	98.80	83.3
20	- 50	263	0.75	15.6	3820	12.7	1240	2.31	73.57	3.78
21	- 53	260	0.75	16.0	3480	13.0	1150	2.27	73.15	3.72
22	- 52	164	1.0	15.4	362	12.6	126	3.08	71.52	3.51
23	- 23	326	1.0	15.8	1950	11.1	262	3.23	90.56	10.6
24	- 21	326	1.0	15.1	1230	9.9	72	3.53	96.16	26.0
25	- 26	314	0.75	14.5	1150	10.5	222	2.63	86.02	7.15
26	- 27	314	0.75	14.8	1060	10.7	282	2.58	80.77	5.20
27	- 29	416	0.75	14.8	245	9.6	12	2.79	96.82	31.4
28	- 75	362	1.0	14.1	2370	7.8	16	4.45	99.63	270
29	- 77	164	1.0	15.9	140	12.9	26	3.07	84.93	6.64
30	- 74	266	1.0	15.1	420	10.4	5	3.61	99.18	122
31	- 73	266	1.0	10.2	2940	6.2	5	5.68	99.90	1000
32	- 71	266	1.0	10.1	2430	5.8	6	5.93	99.86	714
33	- 72	266	0.75	21.8	422	17.3	140	1.75	73.67	3.80
34	- 75	266	0.75	22.3	42	17.6	13	1.72	75.57	4.09

TABLE III

ASSUMED CLEANUP CRITERIA FOR ARGON BLANKET GAS PROCESSING

Feed Gas:	Flow Rate, scfm		10.0
	Composition, mole %;	Argon	99.86
		Krypton	0.004
		Xenon	0.136
	Activity, curies/scf;	Krypton	24.4
		Xenon	542.6
Activity Limit on Gas Returned to Reactor (Absorber Column Off-Gas), curies/scf			0.03

TABLE IV

DESIGN PARAMETERS FOR BLANKET GAS PROCESSING CASE STUDIES

			Case Number		
			1	2	3
Concentrated Product:	Flow, scfm*		0.02	0.10	1.0
	Composition*, %;	Argon	29.07	85.94	98.63
		Krypton	2.06	0.40	0.04
		Xenon	68.87	13.66	1.33
Absorber:	Temperature, °F		- 4	- 4	- 4
	Pressure, atm		40	40	40
	Number of Stages		26	26	26
Fractionator:	Temperature, °F		32	32	84
	Pressure, atm		3	3	7.3
	Number of Stages		26	26	1
Stripper:	Temperature, °F		12	12	12
	Pressure, atm		2	2	2
	Number of Stages		26	26	26
Temperature Assumed for All Condensers, °F			- 4	- 4	- 4

* Solvent-free basis.

component flows are fixed at roughly the same values for all cases by the minimum decontamination factor required*, and the remaining product is simply diluent argon.

The capital costs estimated for the three cases are all shown in table V to be in the neighborhood of $100,000. For comparison purposes, costs estimated for permselective membrane plants designed to the same specifications are also listed[6]. These latter costs, arrived at assuming a membrane cost of $10/sq yd, assembled in stage packages, are seen to be higher than the absorption plant costs.

TABLE V

ESTIMATED CAPITAL COSTS FOR BLANKET GAS TREATMENT SYSTEMS*

	Case Number		
	1	2	3
Concentrated Product Flow Rate, scfm	0.02	0.01	1.0
Absorption Process Cost, $1000	89.4	89.2	96.5
Permselective Membrane Process Cost†, $1000	366	342	289

* The costs shown in this table, for both the absorption and membrane systems, are installed equipment costs and do not include expenses associated with building modifications, etc., required to house the plants. The space requirement for the absorption system is estimated to be about 500 sq ft of floor area (including the control panel space), with a headroom of 30 feet needed for some equipment items. The requirements for the membrane facility should be similarly small. In either case, some spot radiation shielding may be required, depending on overall facility design.

† Rainey, R. H., et al., "Separation of Radioactive Xenon and Krypton from Other Gases by Use of Permselective Membranes", *IAEA Symposium on Operating and Developmental Experience in the treatment of Airborne Radioactive Wastes*, New York, August, 1968 (SM-110/27).

* In the calculations, the minimum decontamination factor was used as a limit for defining acceptable plants rather than as an absolute target to be met exactly.

STATUS AND FUTURE PLANS

During the next report period, the second phase of the pilot plant testing program will be initiated. In these experiments, the performance of refrigerant-11 as the process solvent will be investigated. Also, it is expected that the topical report summarizing the refrigerant-12 work will be completed. It is probable that the conceptual design part of the project will not be actively pursued, unless interest is shown in detailed information for some specific plant case. The optimization studies will, however, be continued.

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