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THERMAL DIFFUSION DEVELOPMENT
DESIGN OF EXPERIMENTS

By

H. L. Brandt
Chemical Engineering Development Operation
Chemical Development Operation
Chemical Research and Development Operation
HANFORD LABORATORIES OPERATION

June 15, 1959

HANFORD ATOMIC PRODUCTS OPERATION

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THERMAL DIFFUSION DEVELOPMENT
DESIGN OF EXPERIMENTS

INTRODUCTION

The Facilities Engineering Operation of the Chemical Processing Department prepared a process study, scope design of a large thermal diffusion plant for xenon isotope separation⁽¹⁾. This scoping was done perforce on the basis of calculations made from exclusively theoretical considerations because actual design data are not available. The designers are of the opinion, however, that such a basis is not adequate to justify the construction of the plant and have, therefore, requested that an appropriate supporting research and development program be carried out. This report presents an experimental plan for obtaining the data required. Anticipated results from the proposed experiments as outlined below, are expected to be useful for determining the correlation of thermal diffusion column theory with practice for this particular system of xenon isotopes. An interpretation of the data will permit the determination of the sensitivity of the column parameters to the change in operational and design variables over which the designer and operator have control. Basic observations made on the behavior of xenon may, in addition, be of general scientific and technological interest. Included in the report are estimates of the kind and quantity of data to be obtained, the analytical services required, and the total analytical and operational cost of the program. The main theoretical concept is given in Part I - Design Parameter Calculations of this report. Two reports, now in preparation, will present the scope design of a pilot unit thermal diffusion apparatus and the auxiliary supporting equipment.

SCOPE

The present work is limited to single stage apparatus for two principal reasons. First, the Chemical Processing Department plant scope design referred to in the Introduction of this report, is of this type and, second, the behavior of the xenon in a single column can be compared with its expected performance on a theoretical basis as well, and at less cost than in the more efficient multiple stage apparatus. The latter would require the fabrication of more than one column. A scrubber, as such, is not included as part of the equipment because it functions, in theory, exactly as does the enricher. Flexibility in design can allow a column to be used either as an enricher or as a scrubber.

COLUMN DIMENSIONS

This experimental program is based on the use of two actual columns, with the following dimensions:

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Column No.	Length (m)	ID	OD	Wall Thickness	
				Outer Tube (cm)	ID
1	6		3.41	4.68	
2	6		11.465	12.735	

NATURE OF PROGRAM REQUIRED

Influence of Column Diameter - The only practical method of obtaining the large rate of production stipulated for the plant, was to use a relatively larger mean gap circumference (B) to obtain the cross sectional area required.

The annular circumference, however, cannot be increased significantly without creating an intolerably large parasitic current factor. Consideration of the ratio of the parasitic to convective transport $[K_p/K_c = (315/16\pi^2)(B/w)^2 (\int T/\Delta T)^2]$ makes clear the implication when the circumference to gap ratio (B/w) is increased. If the annular gap is fixed, an increase in circumference would grossly increase the parasitic transport. Theory and experiment indicated that for most systems a circumference-to-gap ratio of about 20 is desirable.

Experimental Column Design Basis - The phenomenological basis of column design, as given by Furry and Jones, has been confirmed often enough to justify its use as a basis of design. It appears reasonable then to accept their recommended criteria for all column parameters for at least one of the proposed experimental columns, since the objective is to compare the experimental equipment with that of the theoretical. A larger second column may then be made to have a "grey" zone circumference-to-gap ratio (B/2w) of 60 and, yet, have the same annular gap (2w) as the smaller column. This will permit a study of the possibility of increasing the column production rate by increasing the cross-sectional area of the gas chamber, even though this is not in harmony with the theoretical concept given above.

TURBULENT FLOW INVESTIGATION DESIRABLE

An investigation of the effect of turbulence in the xenon gas system can be justified. The fact that the transport equation of Furry and Jones is predicated on a non-turbulent flow imposes a limitation on the value of the thermal diffusion coefficient (H). As a consequence, theoretical calculations will often indicate the use of several columns in parallel, rather than a single column, to attain a larger value of H. Sufficient theoretical and experimental evidence, however, has been presented to question the need for lamellar flow in thermal diffusion column. In the rare gas system A-Ne, for example, it was predicted and confirmed by experiment that in a pressure range of 4 to 108 atmospheres that, even though turbulence occurs over part of the annular gap, a

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separation may be effected and may even be enhanced by this turbulence⁽¹⁾. Other experiments performed in concentric glass tubes with corrugations and baffles to induce turbulence resulted in augmentation of the separation factors (over that of the same tubes without the turbulence inducers) in a mixture of methane and ammonia⁽²⁾. It is possible that some of these techniques introduce a new separations mechanism and do not, therefore, invalidate the Furry and Jones Concept.

The column dimensions are fixed; thus, the annular gap cannot be increased, however, an increase in pressure may be used as a means to increase the K_c/K_d which, it will be recalled, was fixed at 10 in the design. The range of pressure (0.1 to 1.0 atmosphere) in these experiments will permit this ratio to go well over K_c/K_d of 25 (under which conditions the Reynolds number is 10) which is the upper limit for lamellar flow. There is good possibility, therefore, to obtain data which may help resolve the turbulence question.

EXPERIMENTAL PLAN

General - The two experimental columns may be operated simultaneously and probably with the same personnel that would be needed to attend one column. It is estimated from the schedule given below that the experimental work will last five months. The necessity of establishing equilibrium in the thermal diffusion column makes a continuous operation practically mandatory. This means that a three-shift day is required.

Personnel Requirement - The initial shift crew would be: an engineer and an engineering assistant. The day-shift engineer would be in charge. It may be possible, after a given orientation period, for example, two weeks, that the engineering assistant could operate the equipment alone. The operating cost (for two men per shift) based on a recent Chemical Engineering Development "average" of \$46,000 () Per exempt-man year:

3 (engineers) x 0.372 portion of year x \$46,000 = \$51,300 (includes to operate 1-1/2 ⁽³⁾ Engineering assistants)	
1-1/2 x \$6,000 ⁽³⁾ x 0.372	<u>3,350</u>
	\$54,650
add 25% for personnel contingencies	<u>13,660</u>
	\$68,310

Analytical Requirement - Skilled personnel and an adequate mass spectrometer are available in the Analytical Laboratories of Hanford Laboratories Operations

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to absorb the predicted load without augmenting either staff or facility. About 75 analyses a month are required at \$25 + 25 percent for contingencies at a total cost of \$11,400.

Gas Supply - A mixture of xenon isotopes that would be suitable for experimental column feed can be made available by the Atomic Energy Commission⁽⁴⁾.

VARIABLES

Enricher Section

Temperature - A temperature (T) of 879 C is calculated to be necessary to effect the separation specified. It would be desirable to get away from the use of such a high temperature, which is near the practical maximum. Therefore, lower temperature will be tested. See following schedule.

Pressure - The calculated optimum pressure for column operation is 0.4 atmosphere absolute. Higher pressures introduce turbulence according to the basic theory. Nonetheless, a new concept (see above) indicates that turbulence may even enhance separation. The schedule, therefore, includes a pressure range from 1.0 to 0.1 atmosphere.

Product Draw-Off Rate - (σ) The product draw-off rate, enricher length, and enrichment factor are interrelated through the parameters n and y . The σ will be varied by an increment of about 0.2 grams/day (L and P constant) to determine the effect on enrichment.

Enrichment Fiction Length - (L) The effective fiction length of the enricher will be varied by introducing the feed at different points along the column. This parameter will be reduced by increments of 0.5 meters for any given temperature with P and σ constant.

Scrubber - The conformance of the scrubber operation to theory can probably be tested sufficiently while the enrichment section is being tested for a change in length. The length of column above which the feed is introduced may be considered the scrubber. When the feed is introduced at the bottom of the column the enrichment section is nonexistent, thus, the whole column is functioning as a scrubber.

Required
Time
(days)

Function

3

Equipment Shakedown

Heater testing
Water
Rate of flow
Temperature in
Temperature out

Power Requirement
Column evacuated
Radiation column filled with xenon
Radiation and Conduction

3

Assembly of Auxiliary equipment

4

Leak detection

Calibration of volumes, as required,
Including gasometer, ampules, gas lines

Samples*

Shifts (* Estimated number of samples for one column)

12

Feed Sampling

Equilibrium Studies at 0.9 atmosphere absolute
Determine Characteristic time
Plot composition vs. Enricher length

6

8 T₂-879 C

6

8 T₂-800 C

6

8 T₂-750 C

18

24 Repeat above at 0.4 atmosphere absolute
Vary ϕ at given enricher length at T₂ = 879 C
1 for P = 1.0, 0.9, 0.8, 0.7, 0.6, 0.5, 0.4,
0.3, 0.2, 0.1

5

8

5

8

5

8

2 same range of P
3 same range of P

15

24 Repeat as above at T₂ = 800 C

15

24 Repeat as above at T₂ = 750 C

135

72 Change L to L₂ and repeat
72 Change L to L₃ and repeat
72 Change L to L₄ and repeat
10 Scrubber Experiments at 0.4 atmosphere absolute
and a given ϕ .

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SUMMARY

An experimental program is presented for a pilot thermal diffusion apparatus. The objective of the program is to produce design data for plant scale xenon isotope separations equipment. The experiments are designed to determine the correlation of experimental data with those calculated from the existing theory. The two experimental columns have the following dimensions:

<u>Column No</u>	<u>Length (m)</u>	<u>Inner tube OD (cm)</u>	<u>Outer tube ID (cm)</u>
1	6	3.41	4.68
2	6	11.5	12.7

Operational and column variables to be observed include pressure, temperature, rate of production, length of enricher and scrubber sections. In addition to meeting the primary objective, there appears to be good probability for making a contribution to the science and technology of thermal diffusion. The estimated cost of the experimental work is \$80,000. It will require the services of three engineers and three engineering assistants for five months on a 7-day, 3-shift operation.

- NOTE: (1) H. G. Drickamer, et.al. A Modification of the Theory of Thermal Diffusion Columns. J. Chem Phys 18 1945 (1950)
- (2) Brewer, A. and A. Bromley, Journal of Chem. Phys. (1939)
- (3) Switzer, W. O. Personal Communication, April 2, 1959.
- (4) Christy, J. T., AEC-Process Engineering and Manufacturing Division., Hanford Operations Office, Personal Communication.

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