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INTERNAL CORRESPONDENCE

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MASTER

PERFORMANCE TESTS OF THERMAL DIFFUSION COLUMNS
OPERATING WITH METHANE: I. TOTAL REFLUX OPERATION

July 5, 1963

S. Blumkin

INTRODUCTION

The thermal diffusion cascade at the Oak Ridge National Laboratory for separating the carbon isotopes using methane as the process gas has not performed as predicted. The Isotopes Division of ORNL is now about to make a number of experiments to obtain column and cascade separation data to establish a more reliable design basis than the purely theoretical one. The first of these tests is to observe the behavior of a single column operating at total reflux and the second is the behavior of three columns connected in series also operating at total reflux. The theoretical transient behavior of the columns for these two tests has been computed and is presented here to serve as a guide to the experimenters in determining the time intervals at which to take samples and the duration of each test.

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RESULTS

The first test will utilize a nominal 3/4-inch x 15-foot column filled with normal CH₄ and operating at a calorimeter temperature of 400° C. and a pressure of 4 atmospheres with no feed to or withdrawal from the column. Figure 1 presents the calculated C-13 concentration at both ends of the column as a function of time. Since the use of reservoirs at the column ends is likely, curves are presented for several different reservoir sizes. A similar set of curves is given in Figure 2 to represent the identical operation of a cascade of three 7 1/2-foot columns connected in series.

Since methane, free of nitrogen, is evidently not available, the concentration-time gradient for N₂ is also of interest. Figures 3 and 4 show curves of expected N₂ concentrations for the two specified tests, assuming an initial N₂ concentration of 1%.

The steady state concentration gradient for C-13 and N₂ for the three-column test is given in Table I.

DISCUSSION

An operating temperature of 450° C. had been assumed in the design of the cascade to enrich C-13 to a concentration of 90%.* Actual cascade operation showed, however, that operation at this temperature caused fairly rapid clogging of the system as a result of methane decomposition, and the operating temperature was then reduced to about 400° C. For this reason the tests are to be run at 400° C. rather than the original design temperature.

The "pressure swing" method was used in the production cascade to obtain intercolumn transport. It is suspected that this does not provide sufficient transport and that it also results in some mixing losses. The second test should provide a measure of the cascading inefficiency due to this method of intercolumn transport.

The normal methane available contains impurities totaling about 1%. The contaminants consist mostly of nitrogen, small quantities of CO₂ and CO, and traces of higher hydrocarbons such as ethane and propane. The presence of these "heavy" carbon containing impurities has an adverse effect on the C-13 enrichment. When this difficulty was encountered in the production cascade, a charcoal trap was installed in the feed line to the cascade to remove most of these impurities. The methane to be used in these tests will be purified

* S. Blumkin and E. Von Halle, "Cascades for the Enrichment of C-13 to 90% by Thermal Diffusion," Union Carbide Nuclear Company, Oak Ridge Gaseous Diffusion Plant, memorandum to G. A. Garrett, October 18, 1961, KOA-922.

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by means of such a trap. The nitrogen is not removed by this means. The separation factor for N_2 from CH_4 is theoretically almost an order of magnitude larger than that for $C^{13}H_4$. Thus, in a cascade designed for the enrichment of C-13 to 90%, it is necessary to provide a "purge" section to draw off the highly concentrated N_2 . It therefore becomes important also to determine the performance of these columns in separating N_2 from CH_4 .

BASES


The separation parameters on which the calculations were based are summarized in Table II.

The effective heater lengths of the 15-foot and 7 1/2-foot columns are assumed to be 14 feet and 7 feet, respectively.


The value given for α for CH_4 is based on a correlation of experimental data, and is about 84% of the theoretical value.

The separation parameters for N_2 in CH_4 are a function of its concentration, in general increasing with concentration. The values given here are for an N_2 concentration of 1%.

As for the production cascade design, the values given for $K_c + K_d$ for both $C^{13}H_4$ - $C^{12}H_4$ and N_2 - CH_4 separations are 125% of the theoretical values.


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SB:h

TABLE I

STEADY STATE CONCENTRATION GRADIENT IN A SQUARE CASCADE
OF THREE 7 1/2-FOOT COLUMNS OPERATING AT TOTAL REFLUX

Distance from Bottom of Cascade, Columns	Concentration, Mol Fraction					
	$C^{13}H_4$ in CH_4			N_2		
	No Reservoirs	100 cc Reservoirs	1000 cc Reservoirs	No Reservoirs	100 cc Reservoirs	1000 cc Reservoirs
0	0.0367	0.0307	0.0233	0.200	0.0606	0.0250
1	0.0131	0.0105	0.0079	0.11×10^{-4}	0.21×10^{-5}	0.75×10^{-6}
2	0.0049	0.0035	0.0027	0.31×10^{-9}	0.56×10^{-10}	0.20×10^{-10}
3	0.0015	0.0012	0.00095	0.80×10^{-14}	0.15×10^{-14}	0.4×10^{-15}

TABLE II

BASES

Operating Conditions

Temperature	400° C.
Pressure	4 atmospheres

Separation Parameters

	$\frac{C^{13}H_4 - C^{12}H_4}{C^{13}H_4 - C^{12}H_4}$	$\frac{N_2 - CH_4}{N_2 - CH_4}$
α	0.0105	0.0590
H, gm/sec.	0.202×10^{-4}	0.155×10^{-3}
$K_c + K_d$, gm-cm/sec.	0.314×10^{-2}	0.332×10^{-2}
Separative capacity, g./day	0.480	27.0

FIGURE 1
THE ENRICHMENT OF C-13 IN CH₄ AS A FUNCTION OF TIME
IN A 15-FT. COLUMN OPERATING AT TOTAL REFLUX

Parameter: Volume of reservoirs of equal size at
the top and bottom of the column, cc.

Equilibrium Conditions			
Reservoir volume, cc.	C-13 conc. at bottom	C-13 conc. at top	Time to 98% of equil. conc. span, hrs.
0	0.0266	0.00316	10.1
25	0.0255	0.00303	12.7
50	0.0248	0.00294	15.6
100	0.0237	0.00281	21.2
1000	0.0205	0.00243	122.0

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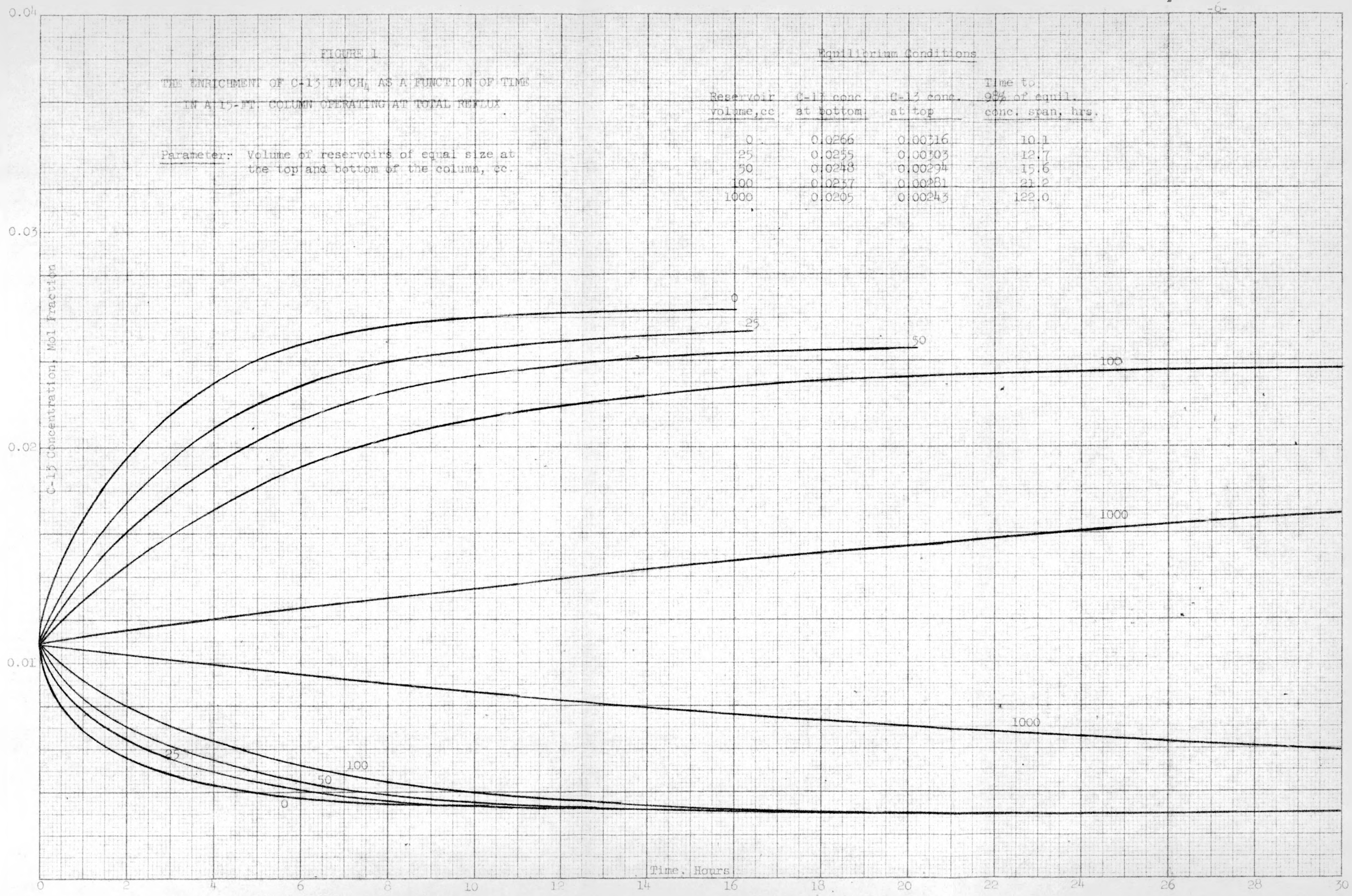


FIGURE 2

THE ENRICHMENT OF C-13 IN CH₄ AS A FUNCTION OF TIME IN A SQUARE CASCADE
OF THREE 7 1/2 FT. COLUMNS OPERATING AT TOTAL REFLEX

Parameter: Volume of reservoirs of equal size at
the top and bottom of the cascade, cc.

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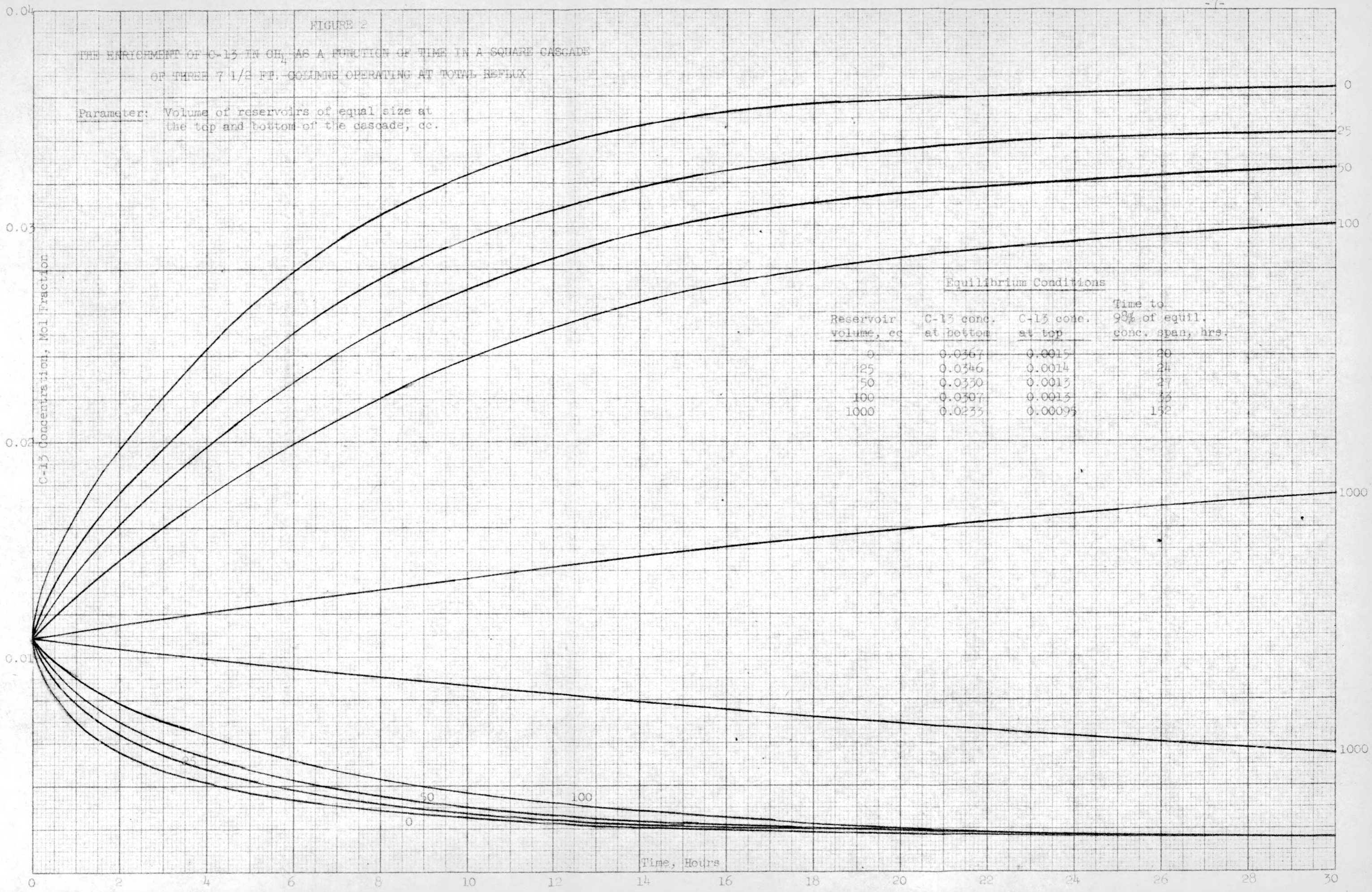


FIGURE 3

N_2-CH_4 SEPARATION AS A FUNCTION OF TIME
IN A 15-FT. COLUMN OPERATING AT TOTAL REFLUX

Parameter: Volume of reservoirs of equal size at the top and bottom of the column, cc.

Equilibrium Conditions

Reservoir Volume, cc	N_2 conc. at bottom	N_2 conc. at top	Time to 98% equil. conc. span, hrs.
0	0.140	0.11×10^{-6}	2.6
25	0.082	0.66×10^{-7}	2.9
50	0.062	0.50×10^{-7}	3.2
100	0.045	0.36×10^{-7}	3.8
1000	0.023	0.18×10^{-7}	21.0

0.15

Bottom N_2 Concentration, Mol Fraction

0.10

0.05

Time, Hours

0

1

2

3

4

FIGURE 4

N_2-CH_4 SEPARATION AS A FUNCTION OF TIME IN A SQUARE CASCADE
OF THREE 7 1/2-FT. COLUMNS OPERATING AT TOTAL REFLUX

Parameter: Volume of reservoirs of equal size at
the top and bottom of the cascade, cc.

