

Copyright Warning & Restrictions

The copyright law of the United States (Title 17, United States Code) governs the making of photocopies or other reproductions of copyrighted material.

Under certain conditions specified in the law, libraries and archives are authorized to furnish a photocopy or other reproduction. One of these specified conditions is that the photocopy or reproduction is not to be “used for any purpose other than private study, scholarship, or research.” If a user makes a request for, or later uses, a photocopy or reproduction for purposes in excess of “fair use” that user may be liable for copyright infringement,

This institution reserves the right to refuse to accept a copying order if, in its judgment, fulfillment of the order would involve violation of copyright law.

Please Note: The author retains the copyright while the New Jersey Institute of Technology reserves the right to distribute this thesis or dissertation

Printing note: If you do not wish to print this page, then select “Pages from: first page # to: last page #” on the print dialog screen

The Van Houten library has removed some of the personal information and all signatures from the approval page and biographical sketches of theses and dissertations in order to protect the identity of NJIT graduates and faculty.

PREDICTIONS OF CATALYST UTILIZATION
WITH CONTINUOUS REPLACEMENT IN MULTISTAGE
CONTINUOUS REACTORS FROM BATCH REACTOR DATA

BY

NORMAN M. FEINBERG

A THESIS

PRESENTED IN PARTIAL FULFILLMENT OF

THE REQUIREMENTS FOR THE DEGREE

OF

MASTER OF SCIENCE IN CHEMICAL ENGINEERING

AT

NEWARK COLLEGE OF ENGINEERING

This thesis is to be used only with due regard to the rights of the author. Bibliographical references may be noted, but passages must not be copied without credit being given in subsequent written or published work.

Newark, New Jersey
1972

ABSTRACT

This thesis presents a new equation for predicting catalyst utilization with continuous replacement in multistage continuous reactors from batch reactor data.

An equation is derived from experimental data for a no-replacement system which predicts catalyst activity as a function of time. The assumption of constant inventory, constant replacement rate and perfect mixing permit the calculation of catalyst age distribution. By combining age distribution with a measure of catalyst productivity, as determined from activity as a function of time, an equation for catalyst utilization was derived. The case used to prepare this equation was where the activity decline curve appears as a straight line on log-log paper.

The resulting equation which describes catalyst utilization is as follows:

$$\text{Catalyst utilization} = \frac{(n-b)\Gamma(n-b)/\Gamma(n)}{r^{i-b}}$$

where: n is the number of reactors in series,

Γ is the gamma function, and

b is the slope of the catalyst activity curve on a log-log plot.

APPROVAL OF THESIS
PREDICTIONS OF CATALYST UTILIZATION WITH CONTINUOUS
REPLACEMENT IN MULTISTAGE CONTINUOUS REACTORS FROM
BATCH REACTOR DATA

BY

NORMAN M. FEINBERG

FOR

DEPARTMENT OF CHEMICAL ENGINEERING
NEWARK COLLEGE OF ENGINEERING

BY

FACULTY COMMITTEE

APPROVAL:

NEWARK, NEW JERSEY

JUNE, 1972

ACKNOWLEDGMENT

The author wishes to express his appreciation to Dr. J. E. McCormick for his helpful assistance and thoughtful review of this work.

TABLE OF CONTENTS

	Page
INTRODUCTION	1
DEVELOPMENT OF THEORRETICAL EQUATION	5
CALCULATION OF CATALYST UTILIZATION	14
DISCUSSION	20
CONCLUSIONS	34
APPENDIX	35

LIST OF FIGURES

Figure		Page
1.	Catalyst Utilization versus Replacement Rate, $b=0.1$	23
2.	Catalyst Utilization versus Replacement Rate, $b=0.2$	24
3.	Catalyst Utilization versus Replacement Rate, $b=0.3$	25
4.	Catalyst Utilization versus Replacement Rate, $b=0.4$	26
5.	Catalyst Utilization versus Slope of No-Replacement Curve, $r=0.01$	27
6.	Catalyst Utilization versus Slope of No-Replacement Curve, $r=0.02$	28
7.	Catalyst Utilization versus Slope of No-Replacement Curve, $r=0.03$	29
8.	Catalyst Utilization versus Slope of No-Replacement Curve, $r=0.04$	30
9.	Catalyst Utilization versus Slope of No-Replacement Curve, $r=0.05$	31
10.	Catalyst Utilization versus Slope of No-Replacement Curve, $r=0.10$	32
11.	Catalyst Utilization versus Number of Stages for Semilog Decline	33

LIST OF TABLES

Table	Page
1. Normgamm Program	15
2. Catalyst Utilization Values	16
A-1. Nomenclature	35
A-2. References	37

INTRODUCTION

The ability to predict the catalyst utilization in a fluidized bed under continuous replacement is of the utmost importance to a design engineer in that field. The money spent for catalyst replacement in a catalytic process is an important contribution to the daily operating cost.

At present, to find out the catalyst utilization for various rates of catalyst replacement, many experimental runs must be made at different rates. This method is time consuming and costly, because each run has to reach steady-state activity. In addition, if a number of reactors in series (staging) is being considered, then runs must also be made to study the effect of such staging (1, 2, 3, etc.).

The intent of this thesis is to develop a theoretical expression for catalyst utilization at any catalyst replacement rate and any given number of stages. Necessary data would come from laboratory and/or pilot plant experiments using equipment such as would be used in a commercial installation but with no replacement of the catalyst. The equation would be developed for the special case of log-log activity decline, based on the equation:

$$y = a t^{-b}$$

Anderson and Matthias have developed equations which predicts catalyst activity for any catalyst replacement

rate from laboratory experiments but with no replacement of the catalyst. They developed equations for three different types of catalyst activity declines, which are (1) linear, (2) experimental, and (3) log-log. A curve for catalyst deactivation, as a function of time, was fitted to experimental data by use of least squares methods. This curve then shows catalyst activity as a function of time under no-replacement conditions. Their proposed prediction equations are only valid, if there is perfect mixing in the reactor and activity of the catalyst in a function time only. Only single stage operations are studied by the above investigators.¹

MacMullin and Weber derived an equation based on the assumptions of constant inventory, constant replacement rate, and perfect mixing, which calculates the fraction of catalyst of age t or younger in a series of equal-volume reactors and also for the special case of a single reactor. By differentiating the above equation, the resultant equation describes the catalyst age distribution.²

The knowledge of the catalyst activity data for the no-replacement system, combined with the steady state age distribution of the catalyst, permits prediction of the steady-state average catalyst activity. The final equation for predicting catalyst activity in a continuous system

is found by mathematical integration of the no-replacement activity curve and catalyst age distribution with respect to time.

Resnick developed equations which predict catalyst utilization when multistage reactors are operated with continuous replacement. He considered three different types of catalyst activity declines, which are (1) linear, (2) experimental, and (3) constant over a finite time interval, and then falling to zero.³

The assumptions Resnick used to develop the equations describing catalyst utilization are as follows:

- (1) Catalyst moves in succession through the reactors.
- (2) Constant inventory is maintained in each reactor, by continuous equal withdrawal and addition of fresh catalyst at constant rate (constant replacement rate).
- (3) Mixing is ideal in that each particle, fresh or old, has the same probability of being withdrawn.
- (4) Activity of the catalyst varies only as a function of time.

By combining the age distribution function, as mentioned previously, with the no-replacement activity curve and mathematically integrating them, Resnick developed an equation which described the yield of product in pounds

4.

per pound of catalyst. Catalyst utilization is then the ratio of yields obtained by continuous replacement and no-replacement operations.

The percent catalyst utilization is then plotted against the coefficients of the catalyst activity equation for a continuous addition with the number of stages as a parameter. By having knowledge of the replacement rate and activity equation coefficients, one can then calculate the percent catalyst utilization for different numbers of reactors.

The above percent catalyst utilization is based on infinite time, but actual operations are not infinite. According to Resnick comparing the catalyst utilization with that for continuous replacement would not be quite fair. Since in real life operations would be for a definite period at which time the reactor(s) would then be dumped and recharged with fresh catalyst. A more justified approach is to evaluate the no-replacement yield at a time equal to the average residence time for continuous replacement. The realistic catalyst utilization would be determined by dividing the percent catalyst utilization based on infinite time, by the yield of product per pound of catalyst at an average catalyst residence time (as defined by the catalyst age distribution) for continuous replacement.

Development of the Theoretical Equation

The assumptions used to develop the catalyst age distribution are as follows:

- (1) Constant inventory is maintained in each reactor
- (2) Constant replacement rate
- (3) Mixing is ideal

The MacMullin and Weber equation based on the above assumptions for the fraction of catalyst of age t or younger in a series of equal-volume reactors is:

$$P = 1 - e^{-rt} \left[1 + rt + \frac{(rt)^2}{2!} + \dots + \frac{(rt)^{n-1}}{(n-1)!} \right] \quad (1)$$

where r = rate of replacement of catalyst, fraction per hour

t = time, hours

P = fraction of catalyst which is age t or less

n = number of equal-volume reactors in series

They differentiated the above equation with respect to time gives the catalyst age distribution:

$$p = \frac{dp}{dt} = \frac{re^{-rt}(rt)^{n-1}}{(n-1)!} \quad (2)$$

where p = fraction of catalyst which is age t

(probability density fraction for age of catalyst)

The above development by MacMullin and Weber is a general equation independent of the catalyst activity decline.

By combining the catalyst age distribution with a measure of catalyst productivity as determined from catalyst activity decline, as a fraction of time, an equation for catalyst utilization can be derived.

The case used in developing this thesis corresponded to a real operational approach. This assumes that activity decays according to Equation 3 which is the case of a so-called log-log decline of activity.

$$y = at^{-b} \quad (3)$$

where y = catalyst activity index

t = time, hours

a, b = least squares constants

A measure of the cumulative productivity for catalyst of age t is obtained by integrating Equation 3 over the time interval 0 to t :

$$Y_t = \int_0^t at^b = \frac{a}{(1-b)} t^{(1-b)} \quad (4)$$

where Y_t = cumulative productivity, (total yield of product for catalyst to time t), pounds of product per pound of catalyst

Combining Equations 2 and 4 and integrating gives the total yield for continuous replacement. For one reactor, this yield, in pounds of product per pound of catalyst, would be:

$$Y_{r1} = \int_0^{\infty} \frac{a}{(1-b)} (t^{1-b}) r e^{-rt} dt$$

To evaluate this integral, we let $b=2-\alpha$, and therefore $\alpha=2-b$. From this: $(1-b)=\alpha-1$

$$Y_{r1} = \int_0^{\infty} \frac{ar}{(\alpha-1)} t^{\alpha-1} e^{-rt} dt \cdot \frac{r^{\alpha}}{r^{\alpha}} = \int_0^{\infty} \frac{ar^{\alpha}}{(\alpha-1)r^{\alpha-1}} t^{\alpha-1} e^{-rt} dt$$

Integration of this equation requires use of Euler's integral

$$\int_0^{\infty} r^{\alpha} x^{\alpha-1} e^{-rx} dx = \Gamma(\alpha) \frac{1}{r^{\alpha}},$$

thus

$$Y_{r1} = \frac{a}{(\alpha-1)r^{\alpha-1}} \Gamma(\alpha) = \frac{ar}{(1-b)} \frac{1}{r^{2-b}} \Gamma(2-b)$$

and using the fact that

$$\Gamma(2-b) = (1-b) \Gamma(1-b)$$

we find

$$Y_{r_1} = \frac{a}{r^{1-b}} \Gamma(1-b) \quad (5)$$

For the case with n reactors in series,

$$Y_{r_n} = \int_0^{\infty} \frac{a}{(1-b)} [t^{1-b}] \frac{r e^{-rt} (rt)^{n-1}}{(n-1)!} dt$$

letting $b=1-\beta$ or $\beta=1-b$

$$\begin{aligned} Y_{r_n} &= \int_0^{\infty} \frac{a}{(1-b)} t^{n-b} \frac{r^n e^{-rt}}{(n-1)!} dt \cdot \frac{r^\beta}{r^\beta} \\ &= \int_0^{\infty} \frac{a}{(1-b)r^\beta} t^{(n+\beta-1)} \frac{r^{n+\beta}}{(n-1)!} e^{-rt} dt \\ &= \frac{a}{(1-b)r^\beta} \frac{\Gamma(n+\beta)}{(n-1)!} \end{aligned}$$

and finally

$$Y_{r_n} = \frac{a(n-b) \int_0^n (n-b)}{(1-b)r^{1-b} \int_0^n (n)} \quad (6)$$

This equation calculates the catalyst utilization.

Catalyst utilization is defined as the ratio of yields obtained for continuous replacement and no-replacement operation. Therefore one must calculate the ultimate yield.

$$\begin{aligned} Y_u &= \int_0^\infty at^{-b} dt \\ &= \int_0^1 at^{-b} dt + \int_1^\infty at^{-b} dt \\ &= \frac{a}{1-b} \left[t^{1-b} \Big|_0^1 - 1 \right] \end{aligned}$$

At $t=0$, Y does not have a value, but at some time $t=c$, after time, $t=0$, Y has a value which is:

$$Y_u = \left[\frac{a}{1-b} \right] (K) \quad (7)$$

The assumed value of Y is expressed in the form above, to enable one to derive the catalyst utilization without a dependence on the constant a .

Since, Y now has a value at time near zero, one must enter this value in Equation 4 and rederive Equations 5 and 6. Proceeding as before we get:

$$Y_t = \left(\frac{a}{1-b} \right) \left[t^{1-b} + K \right] \quad (8)$$

Combining Equations 2 and 8 and integrating gives the total yield for continuous replacement. For one reactor, this yield would be:

$$\begin{aligned} Y_{r_1} &= \int_0^{\infty} \frac{a}{1-b} (t^{1-b} + K) r e^{-rt} dt \\ &= \frac{a}{1-b} \left[\int_0^{\infty} r t^{1-b} e^{-rt} dt + \int_0^{\infty} K r e^{-rt} dt \right] \\ &= \frac{a}{(1-b)} \left[\frac{(1-b) \Gamma(1-b)}{r^{1-b}} + K \right] \\ Y_{r_1} &= \frac{a}{(1-b)} \left[\frac{(1-b) \Gamma(1-b) + K r^{1-b}}{r^{1-b}} \right] \end{aligned}$$

While for n reactors in series,

$$Y_{r_n} = \int_0^{\infty} \frac{a}{(1-b)} \left(t^{1-b+K} \right) \frac{r e^{-rt} (rt)^{n-1}}{(n-1)!} dt$$

$$= \frac{a}{(1-b)(n-1)!} \left[\int_0^{\infty} t^{n-b} r^n e^{-rt} dt + \int_0^{\infty} K t^{n-1} r^n e^{-rt} dt \right]$$

Letting $b=1-\beta$, and $\beta=1-b$ we get

$$Y_{r_n} = \frac{a}{(1-b)(n-1)!} \left[\int_0^{\infty} t^{(n+\beta-1)} r^n e^{-rt} dt \cdot \frac{r^\beta}{r^\beta} + \int_0^{\infty} K t^{n-1} r^n e^{-rt} dt \right]$$

rearranging terms:

$$Y_{r_n} = \frac{a}{(1-b)(n-1)!} \left[\int_0^{\infty} t^{(n+\beta-1)} \frac{r^{n+\beta}}{r^\beta} e^{-rt} dt + \int_0^{\infty} K t^{n-1} r^n e^{-rt} dt \right]$$

Using Euler's Integrals

$$\Gamma(n+\beta) = \int_0^{\infty} r^{n+\beta} t^{(n+\beta-1)} e^{-rt} dt, \quad \text{and}$$

$$\Gamma(n) = \int_0^{\infty} r^n t^{n-1} e^{-rt} dt$$

we integrate the above equation and obtain the following equation:

$$\begin{aligned}
 Y_{r_n} &= \frac{a}{(1-b)(n-1)!} \left[\frac{\Gamma(n+1)}{r^e} + K \Gamma(n) \right] \\
 &= \frac{a}{(1-b)(n-1)!} \left[\frac{\Gamma(n-b+1)}{r^{1-b}} + K \Gamma(n) \right] \\
 Y_{r_n} &= \frac{a}{(1-b)r^{1-b}} \left[\frac{(n-b)\Gamma(n-b)}{\Gamma(n)} + K r^{1-b} \right] \quad (9)
 \end{aligned}$$

Catalyst utilization is the ratio of Equations 8 and 9:

$$R_{I_u} = \frac{(n-b)\Gamma(n-b)/\Gamma(n)}{K r^{1-b}} + 1 \quad (10)$$

where R_{I_u} = catalyst utilization based on no-replacement yield and infinite time.

One can now calculate a catalyst utilization, based on removing the K term from the left side of the equation.

$$R_{I_u} = (R_{I_u} - 1) K = \frac{(n-b)\Gamma(n-b)/\Gamma(n)}{r^{1-b}} \quad (11)$$

The above equation is just as valid as Equation 10, because when a log-log activity-time plot is assumed, nobody really cares what the value of Y is near zero time. The only concern is that the curve is linear on log-log paper after some initial lag time. This Equation 11 permits calculation of catalyst utilization for n reactors and requires minimal experimental data. The equation reduces to

$$\Gamma(2-b)/r^{1-b}$$

for the special case $n=1$.

Calculation of Catalyst Utilization

The program used to calculate catalyst utilization values from Equation 11 is shown in Table 1. This program makes use of IBM basic language and is called Normgamm. Normgamm calculates catalyst utilization, much as one would do so by hand, that is starts at $r=0.01$ and for all values of $b(0.1$ to $0.7)$ and $n(1$ to $10)$ and calculates catalyst utilization. Gamma function values are calculated by calling a gamma subprogram and then entering this value in Equation 11. Lines 270 to 650 in the Normgamm program are a standard subroutine by IBM to calculate gamma functions. The program then calculates more catalyst utilization numbers for r from 0.01 to 0.10 , in steps of 0.01 as mentioned above.

Table 2 summarizes the catalyst utilization values for b from 0.1 to 0.7 , for n from 1 to 10 and for r from 0.01 to 0.10 which were calculated by the Normgamm program.

When the slope (b) of the catalyst activity decline has been chosen and/or calculated from experimental data, the catalyst utilization can be calculated for different n and r by narrowing down the range of b values used in the Normgamm program.

NORMGAMM 09:56 01/06/72 THURSDAY 101

```

100: #####.### #####.## #####.## #####.## #####.## #####.##
110 FOR r = .01 TO .1 STEP .01
120 PRINT USING 100,r
130 PRINT
140 FOR b = .1 TO .8 STEP .1
150 FOR n = 1 TO 10
160 Z1=(n - b+1)
170 GO SUB 270
180 B1 = G1
190 Z1=N
200 GO SUB 270
210 B2=G1
220 C(n)=B1/B2*(1/r*(1-b))
221 IF C(n)< 100 GO TO 230
222 C(n)=100
230 NEXT n
240 PRINT USING 100 b, C(1), C(2), C(3), C(4), C(5), C(6), C(7), C(8), C(9), C(10)
250 NEXT b
260 NEXT r
265 GO TO 660
270 Z=Z1
280 I=0
290 IF ABS(Z)>1E-6 GO TO 370
300 I=1
310 PRINT USING 360
320 G1=0
330 G=G1
340 Z1=Z
350 GO TO 650
360: ARGUMENT IS WITHIN .000001 OF ZERO OR IS A NEGATIVE INTEGER.
370 A5=1E-6
380 G=1
390 G1=1
400 G2=1
410 IF Z1>2 GO TO 460
420 IF Z1=2 GO TO 560
430 IF Z1<2 GO TO 500
440 IF Z1=2 GO TO 560
450 IF Z1<2 GO TO 520
460 Z1=Z1-1
470 G2=G2*Z1
480 G1=G2
490 GO TO 440
500 IF Z1=1 GO TO 560
510 IF Z1<1 GO TO 570
520 Y=Z1-1
530 G1=-.5684729+Y*(.2548205+Y*(-.0514993))
540 G3=1+Y*(-.5771017+Y*(.985854+Y*(-.8764218+Y*(.8328212+Y*G1))))
550 G1=G2*G3
560 GO TO 330
570 D1=INT(Z1)-Z1
580 IF ABS(D1)≤A5 GO TO 300
590 IF (1-ABS(D1)-A5)≤0 GO TO 300
600 IF Z1>1 GO TO 520
610 G2=G2/Z1
620 Z1=Z1+1
630 G1=G2
640 GO TO 600
650 RETURN
660 END

```


TABLE 2 (1 of 4)

Catalyst Utilization Values

RUN						
NORMGAMM 09:58 01/06/72 THURSDAY 101						
r =	n =	1	2	3	4	5
0.010	b = 0.100	60.68	100.00	100.00	100.00	100.00
	0.200	37.08	66.74	93.44	100.00	100.00
	0.300	22.82	38.80	52.38	64.60	75.91
	0.400	14.16	22.66	29.46	35.35	40.65
	0.500	8.86	13.29	16.62	19.39	21.81
	0.600	5.60	7.84	9.41	10.66	11.72
	0.700	3.57	4.64	5.34	5.88	6.32
0.020	b = 0.100	32.52	61.79	89.59	100.00	100.00
	0.200	21.30	38.33	53.67	67.98	81.57
	0.300	14.05	23.88	32.24	39.77	46.73
	0.400	9.34	14.95	19.43	23.32	26.82
	0.500	6.27	9.40	11.75	13.71	15.42
	0.600	4.24	5.94	7.13	8.08	8.89
	0.700	2.90	3.77	4.34	4.77	5.13
0.030	b = 0.100	22.58	42.90	62.20	80.36	99.05
	0.200	15.40	27.71	38.80	49.15	58.98
	0.300	10.58	17.98	24.28	29.94	35.18
	0.400	7.33	11.72	15.24	18.28	21.03
	0.500	5.12	7.67	9.59	11.19	12.59
	0.600	3.61	5.05	6.00	6.87	7.56
	0.700	2.57	3.34	3.84	4.23	4.54
0.040	b = 0.100	17.43	33.11	48.01	62.41	76.46
	0.200	12.23	22.02	30.82	39.04	46.85
	0.300	8.65	14.70	19.85	24.48	28.76
	0.400	6.16	9.86	12.82	15.39	17.69
	0.500	4.43	6.65	8.31	9.69	10.90
	0.600	3.22	4.50	5.40	6.12	6.73
	0.700	2.36	3.06	3.52	3.88	4.17
0.050	b = 0.100	14.26	27.09	39.28	51.06	62.55
	0.200	10.23	18.42	25.78	32.66	39.19
	0.300	7.40	12.58	16.93	20.94	24.60
	0.400	5.39	8.63	11.21	13.46	15.48
	0.500	3.96	5.94	7.43	8.67	9.75
	0.600	2.94	4.12	4.94	5.60	6.16
	0.700	2.20	2.87	3.30	3.63	3.90

TABLE 2 (2 of 4)

Catalyst Utilization Values

r = 0.010						
	n = 6	7	8	9	10	
b = 0.100	100.00	100.00	100.00	100.00	100.00	100.00
0.200	100.00	100.00	100.00	100.00	100.00	100.00
0.300	86.54	96.63	100.00	100.00	100.00	100.00
0.400	45.53	50.08	54.37	58.45	62.35	62.35
0.500	23.99	25.99	27.85	29.59	31.23	31.23
0.600	12.66	13.51	14.28	14.99	15.66	15.66
0.700	6.70	7.03	7.33	7.61	7.86	7.86
r = 0.020						
b = 0.100	100.00	100.00	100.00	100.00	100.00	100.00
0.200	94.62	100.00	100.00	100.00	100.00	100.00
0.300	53.27	59.48	65.43	71.16	76.69	76.69
0.400	30.04	33.04	35.87	38.56	41.13	41.13
0.500	16.96	18.38	19.69	20.92	22.08	22.08
0.600	9.60	10.24	10.82	11.36	11.87	11.87
0.700	5.44	5.71	5.95	6.18	6.38	6.38
r = 0.030						
b = 0.100	100.00	100.00	100.00	100.00	100.00	100.00
0.200	63.41	77.53	86.39	95.03	100.00	100.00
0.300	40.11	44.79	49.26	53.57	57.74	57.74
0.400	23.55	25.90	28.13	30.23	32.25	32.25
0.500	13.85	15.01	16.03	17.03	18.03	18.03
0.600	8.16	8.70	9.20	9.66	10.09	10.09
0.700	4.82	5.06	5.27	5.47	5.65	5.65
r = 0.040						
b = 0.100	90.22	100.00	100.00	100.00	100.00	100.00
0.200	54.35	61.59	68.63	75.50	82.21	82.21
0.300	32.79	36.62	40.28	43.80	47.21	47.21
0.400	19.82	21.80	23.67	25.44	27.14	27.14
0.500	12.00	12.99	13.92	14.79	15.61	15.61
0.600	7.27	7.76	8.20	8.61	8.99	8.99
0.700	4.42	4.64	4.84	5.02	5.19	5.19
r = 0.050						
b = 0.100	73.80	84.87	95.79	100.00	100.00	100.00
0.200	45.46	51.52	57.41	63.15	68.77	68.77
0.300	28.05	31.32	34.45	37.47	40.38	40.38
0.400	17.33	19.07	20.70	22.25	23.74	23.74
0.500	10.73	11.62	12.45	13.23	13.97	13.97
0.600	6.65	7.10	7.50	7.88	8.23	8.23
0.700	4.13	4.34	4.52	4.69	4.85	4.85

Catalyst Utilization Values

TABLE 2 (3 of 4)

10.

r =		n = 1	2	3	4	5	
0.060	b =	0.100	12.10	22.99	33.33	43.33	53.08
		0.200	8.84	15.92	22.23	28.23	33.87
		0.300	6.51	11.07	14.94	18.43	21.66
		0.400	4.83	7.73	10.05	12.06	13.87
		0.500	3.62	5.43	6.78	7.91	8.90
		0.600	2.73	3.83	4.59	5.21	5.73
		0.700	2.09	2.71	3.12	3.43	3.69
0.070	b =	0.100	10.53	20.01	29.01	37.72	46.20
		0.200	7.82	14.07	19.70	24.95	29.94
		0.300	5.85	9.94	13.42	16.55	19.44
		0.400	4.41	7.05	9.16	11.00	12.65
		0.500	3.35	5.02	6.28	7.33	8.24
		0.600	2.57	3.60	4.32	4.89	5.38
		0.700	1.99	2.59	2.98	3.28	3.52
0.080	b =	0.100	9.34	17.74	25.73	33.45	40.97
		0.200	7.03	12.65	17.70	22.42	26.91
		0.300	5.32	9.05	12.22	15.07	17.71
		0.400	4.07	6.51	8.46	10.15	11.67
		0.500	3.13	4.70	5.87	6.85	7.71
		0.600	2.44	3.41	4.09	4.64	5.10
		0.700	1.91	2.49	2.86	3.15	3.38
0.090	b =	0.100	8.40	15.96	23.14	30.08	36.85
		0.200	6.39	11.51	16.11	20.41	24.49
		0.300	4.90	8.33	11.25	13.88	16.31
		0.400	3.79	6.06	7.88	9.46	10.88
		0.500	2.95	4.43	5.54	6.46	7.27
		0.600	2.32	3.25	3.91	4.43	4.87
		0.700	1.85	2.40	2.76	3.04	3.27
0.100	b =	0.100	7.64	14.52	21.05	27.36	33.52
		0.200	5.88	10.58	14.81	18.76	22.51
		0.300	4.55	7.74	10.45	12.89	15.15
		0.400	3.56	5.69	7.40	8.88	10.21
		0.500	2.80	4.20	5.25	6.13	6.90
		0.600	2.23	3.12	3.74	4.24	4.67
		0.700	1.79	2.33	2.68	2.94	3.17

Catalyst Utilization Values

19.

TABLE 2 (4 of 4)

 $r = 0.060$

	n = 6	7	8	9	10
b = 0.100	62.63	72.02	81.29	90.44	99.48
0.200	39.29	44.53	49.62	54.58	59.43
0.300	24.69	27.57	30.33	32.98	35.54
0.400	15.54	17.09	18.56	19.95	21.28
0.500	9.79	10.61	11.37	12.08	12.75
0.600	6.18	6.60	6.97	7.32	7.65
0.700	3.91	4.11	4.23	4.44	4.59

 $r = 0.070$

b = 0.100	54.52	62.70	70.76	78.72	86.59
0.200	34.73	39.36	43.86	48.25	52.54
0.300	22.16	24.75	27.22	29.61	31.91
0.400	14.16	15.58	16.92	18.19	19.40
0.500	9.07	9.82	10.52	11.18	11.80
0.600	5.81	6.20	6.56	6.88	7.19
0.700	3.73	3.92	4.09	4.24	4.38

 $r = 0.080$

b = 0.100	48.35	55.60	62.75	69.81	76.79
0.200	31.21	35.38	39.42	43.36	47.22
0.300	20.19	22.54	24.79	26.96	29.06
0.400	13.07	14.33	15.61	16.79	17.90
0.500	8.48	9.19	9.84	10.46	11.04
0.600	5.51	5.88	6.22	6.53	6.82
0.700	3.59	3.77	3.93	4.08	4.21

 $r = 0.090$

b = 0.100	43.48	50.01	56.44	62.79	69.06
0.200	28.41	32.20	35.87	39.46	42.97
0.300	18.59	20.76	22.83	24.83	26.76
0.400	12.18	13.40	14.55	15.64	16.68
0.500	8.00	8.66	9.28	9.86	10.41
0.600	5.26	5.61	5.93	6.23	6.50
0.700	3.46	3.64	3.79	3.93	4.07

 $r = 0.010$

b = 0.100	39.55	45.48	51.33	57.11	62.82
0.200	26.11	29.59	32.97	36.27	39.50
0.300	17.27	19.28	21.21	23.06	24.86
0.400	11.44	12.58	13.66	14.68	15.66
0.500	7.59	8.22	8.81	9.36	9.88
0.600	5.04	5.38	5.68	5.97	6.23
0.700	3.36	3.52	3.67	3.81	3.94

Discussion

Figures 1, 2, 3, and 4 show plots of catalyst utilization as a function of r (the replacement rate) with n (number of stages) as a parameter with constant values of b (0.1, 0.2, 0.3, and 0.4).

It appears from the forementioned figures that as r increases, at any value of b (slope) the percent improvement in catalyst utilization due to staging appears to be about the same. But, of course, the absolute value of catalyst utilization is decreasing as would be expected. Also as b increases from 0.1 to 0.4, the curves due to n (the number of stages) squeeze together, which implies that the effect of staging diminishes as the value of the slope increases.

Figures 5, 6, 7, 8, 9, and 10 show plots of catalyst utilization as a function of b (slope) with n (number of stages) as a parameter with constant values of r (0.01, 0.02, 0.03, 0.04, 0.05, and 0.10).

From the above figures, one can see that as b increases, at any value of r (the replacement rate) the percent improvement in catalyst utilization due to staging decreases and at the same time also the absolute value of the catalyst utilization decreases. At larger values of

b, the increase in catalyst utilization due to staging decreases to almost very little improvement.

To sum up, the highest catalyst utilization is achieved at the lowest replacement rate and with the lowest slope.

Figure 11 shows a plot of catalyst utilization as a function of n (the number of stages) with the ratio of $F/(b+r)$ as a parameter. This plot is for a semi-log decline in catalyst activity and shows more clearly than Resnick the effect of staging.

In Resnick's example for a case of where $b=0.1$, $n=1$, and $r=0.05$, the catalyst utilization is 66.7%. But when he compares this case to a two stage case, he then changes r to be 0.10. r should stay the same and the only difference is that the number of stages are increased. For two stage case, the catalyst utilization should be about 85% rather than 75% as shown by Resnick.

For one-stage system where $b=0.1$, and $r=0.05$, the catalyst utilization for a log-log decline would be 14%. For two-stage system, the catalyst utilization would be 27%.

For the above case the effect of staging for a log-log decline versus a semi-log decline results in a

greater increase in the catalyst utilization (93% versus 27.5%), but it must be noted that the catalyst utilization for the log-log case has the greatest room for improvement. The type of activity decline is shown by the above case to be of the utmost importance.

FIGURE 1

CATALYST UTILIZATION
VERSUS
REPLACEMENT RATE

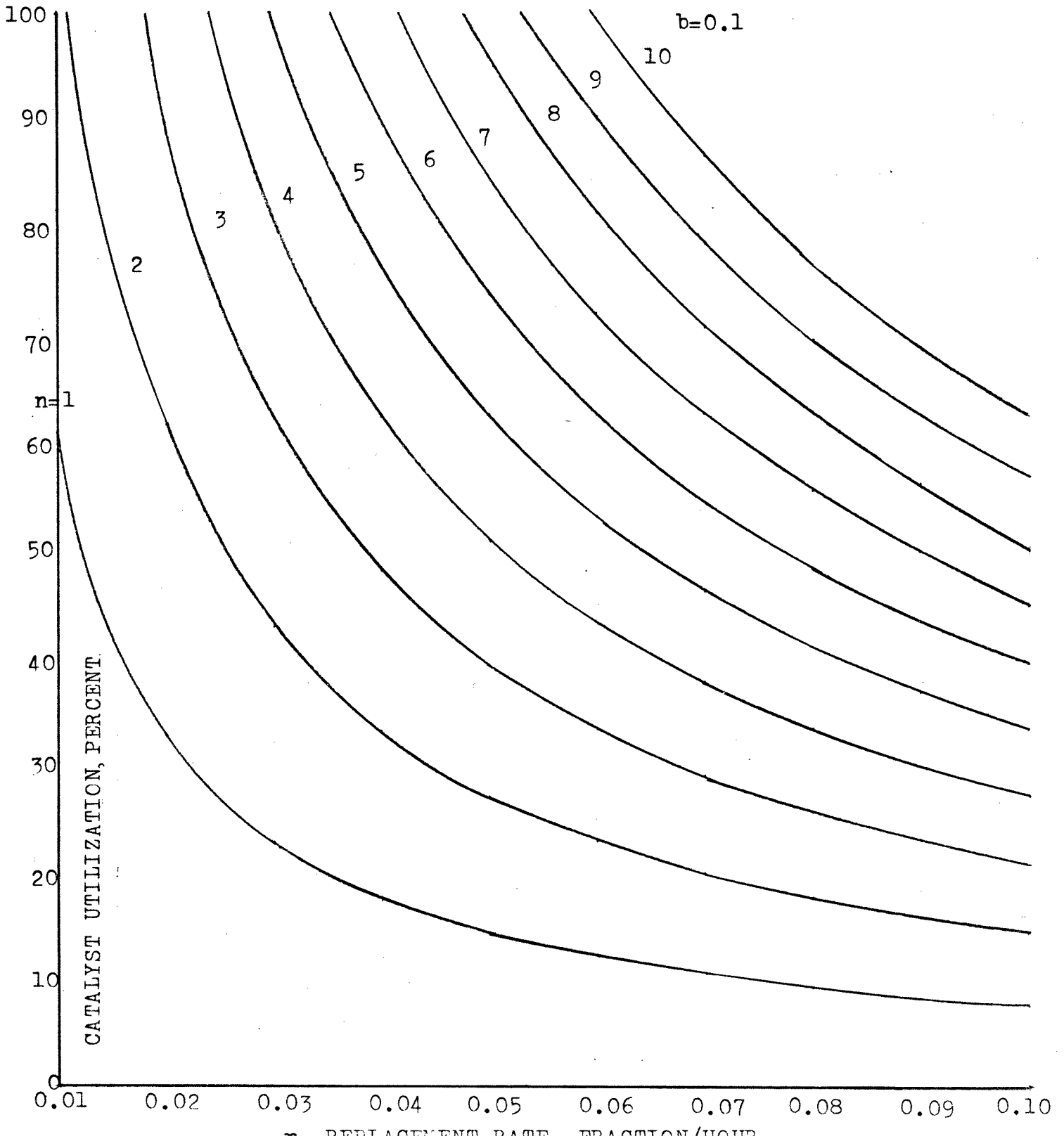


FIGURE 2

CATALYST UTILIZATION
VERSUS
REPLACEMENT RATE

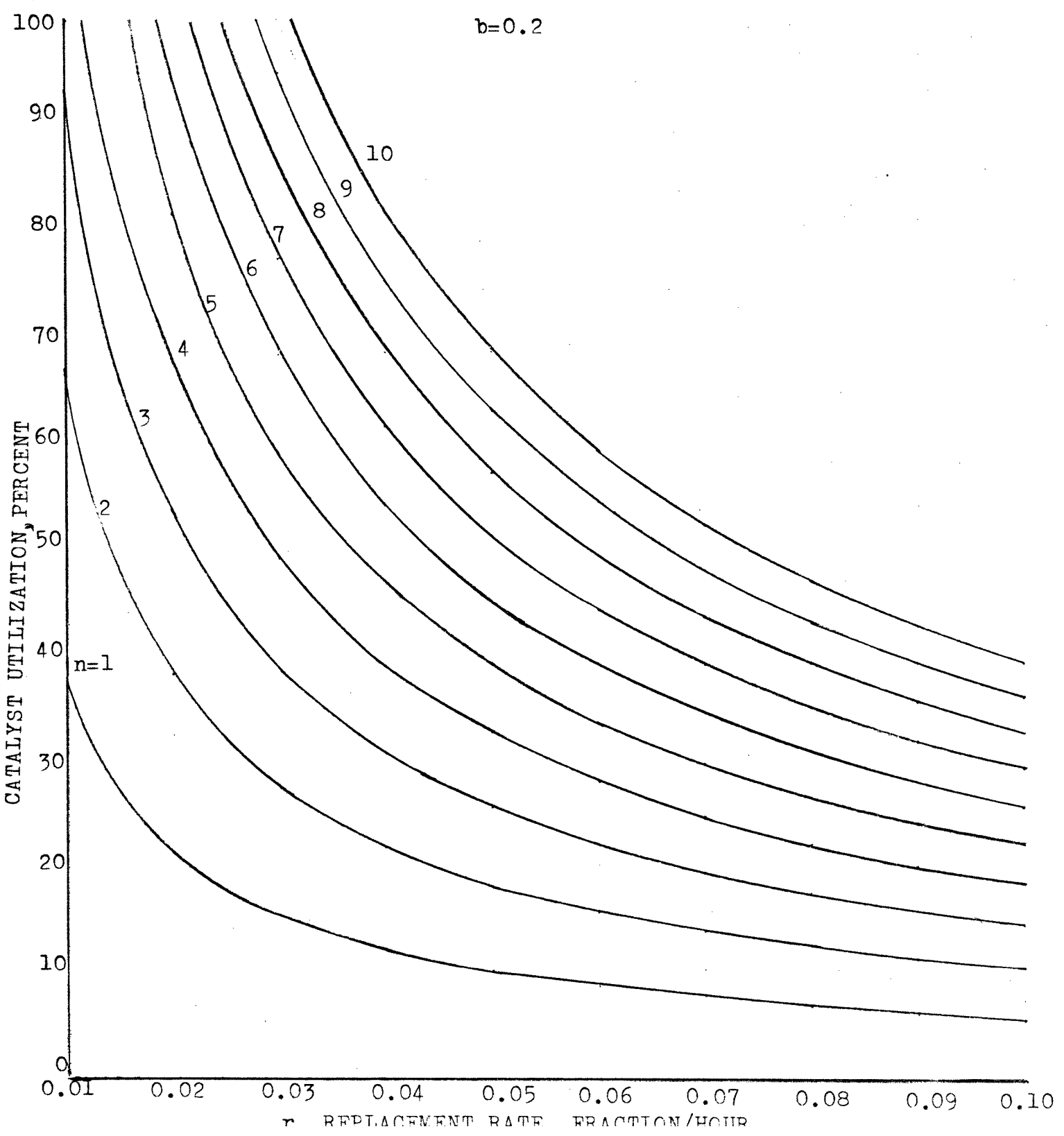


FIGURE 3

CATALYST UTILIZATION
VERSUS
REPLACEMENT RATE

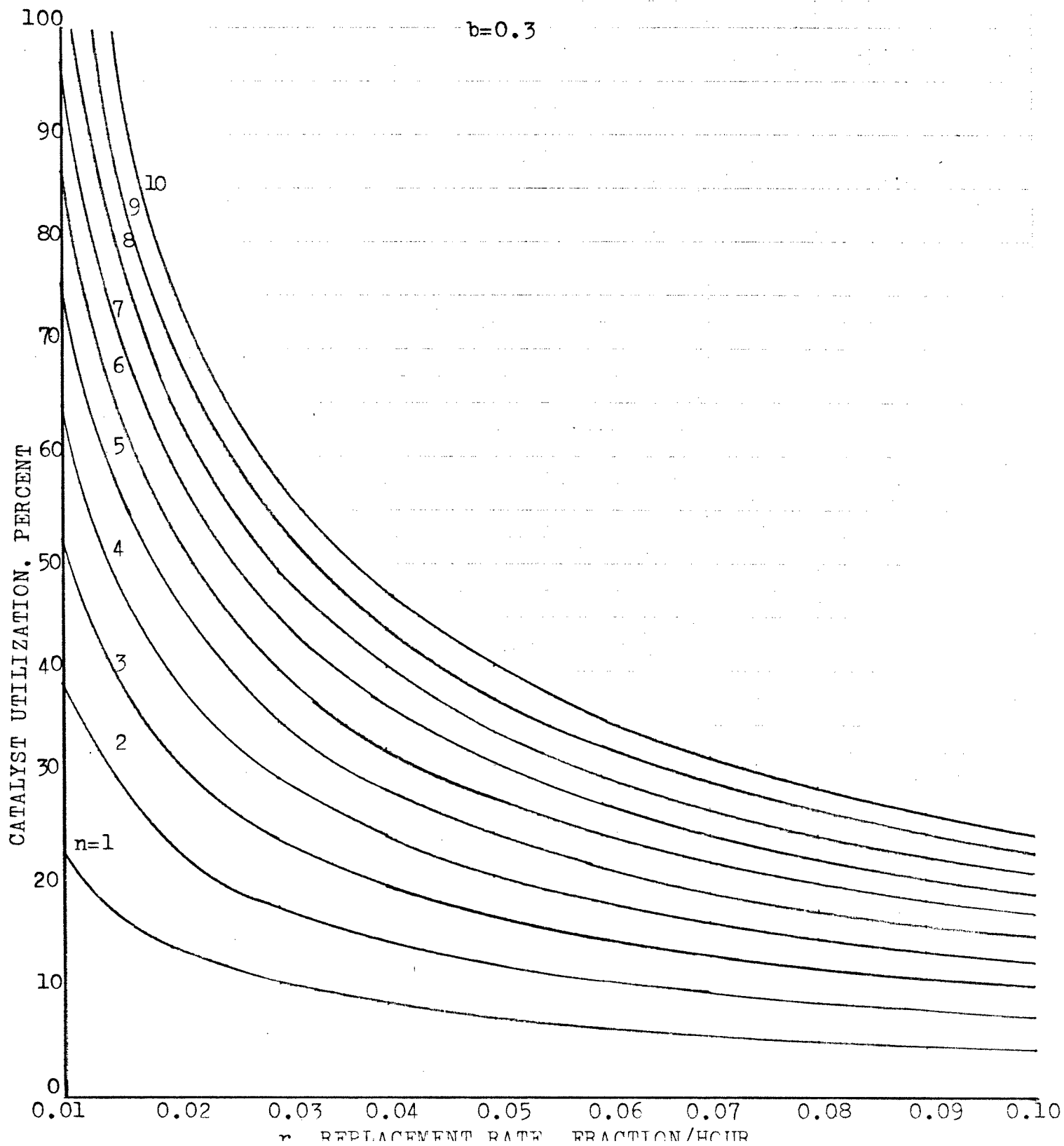


FIGURE 4
CATALYST UTILIZATION
VERSUS
REPLACEMENT RATE

$b=0.4$

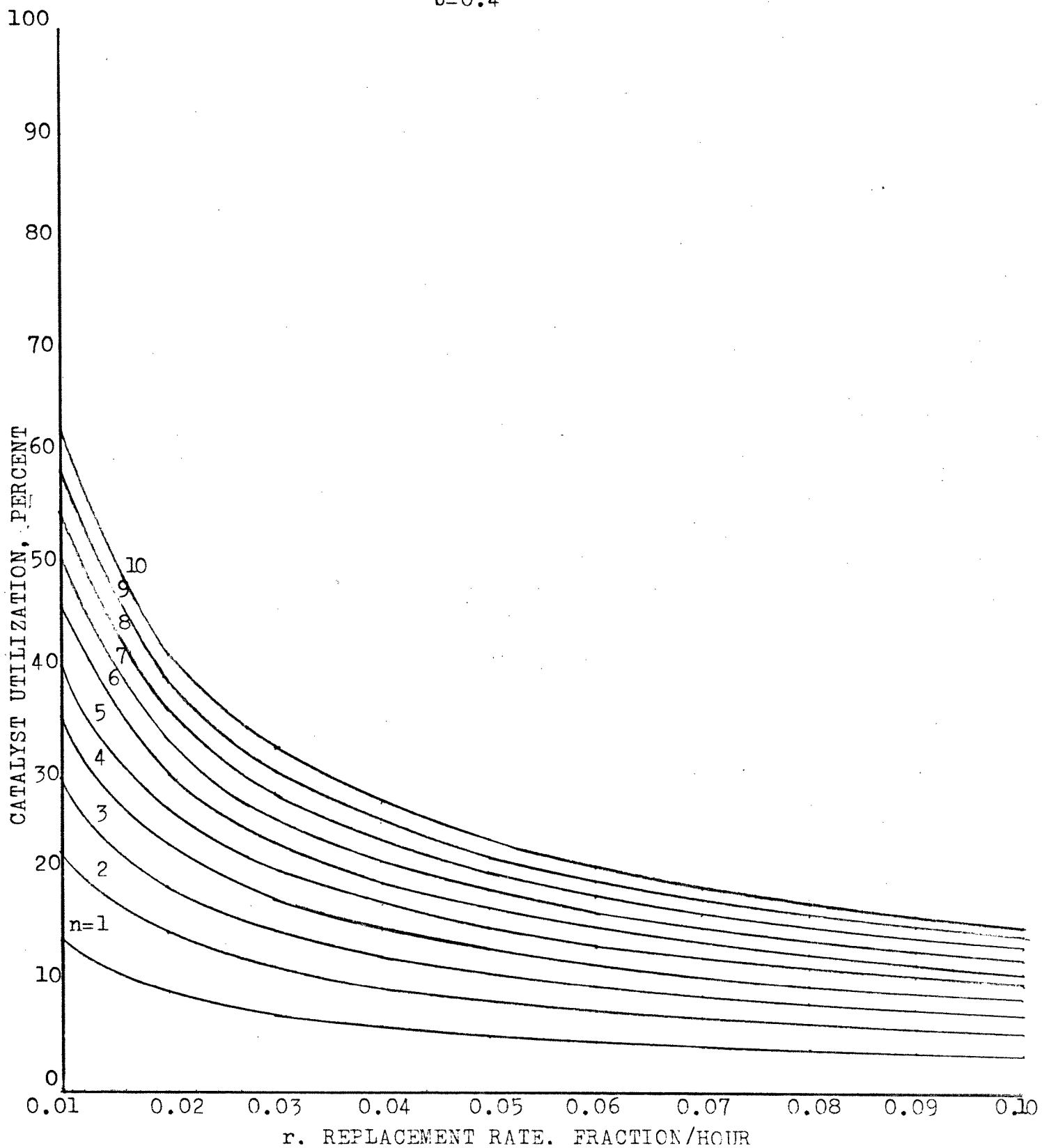


FIGURE 5

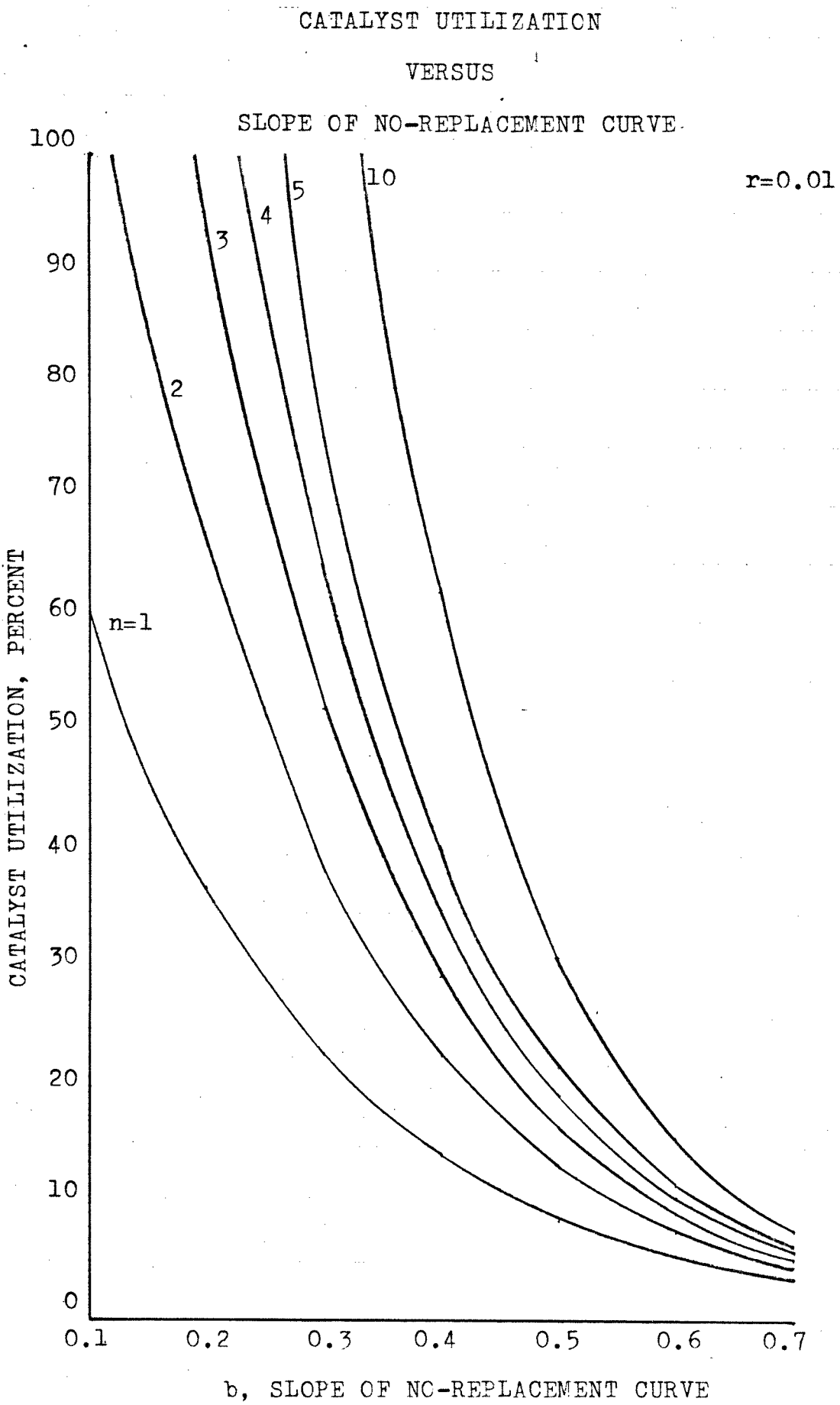


FIGURE 6

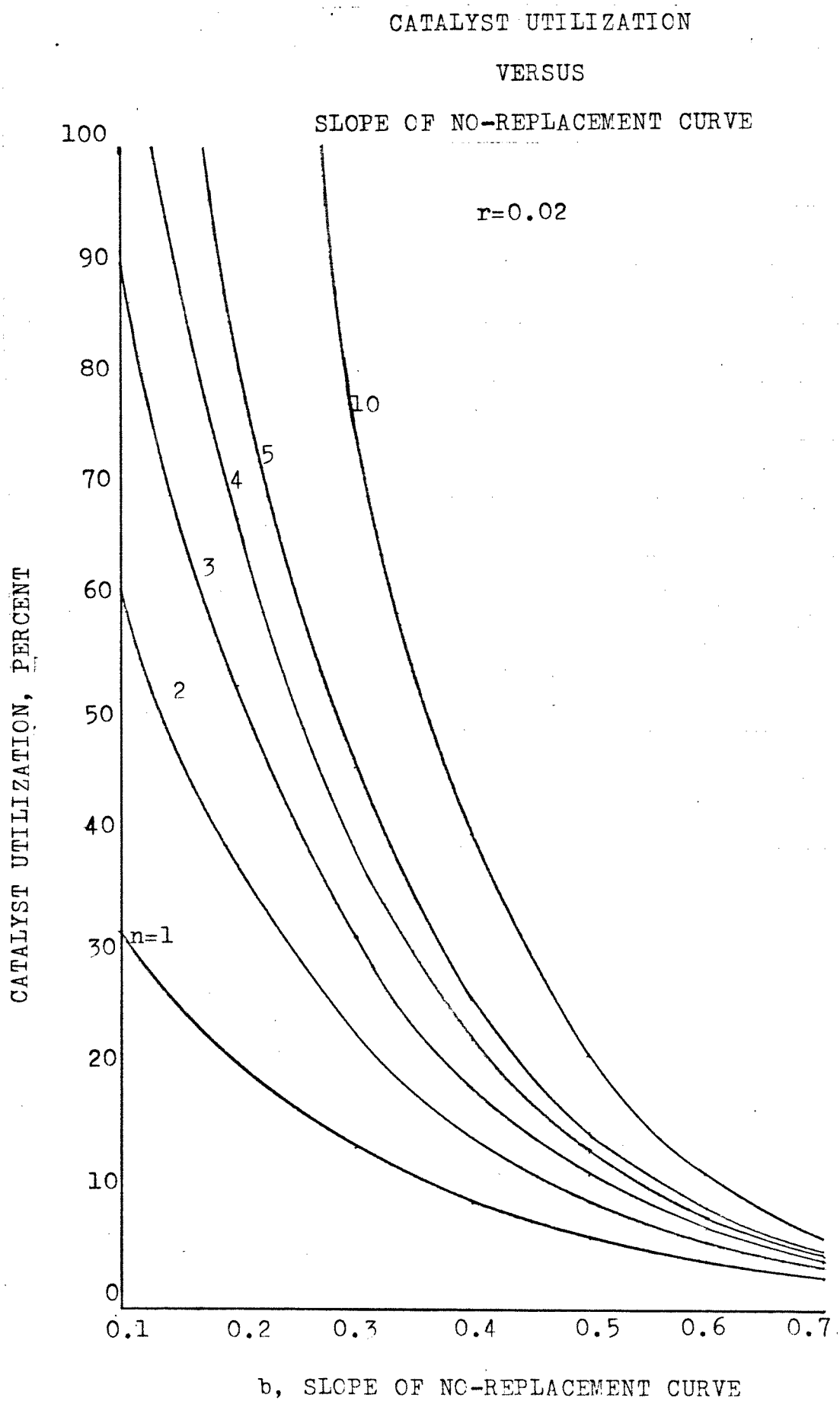


FIGURE 7

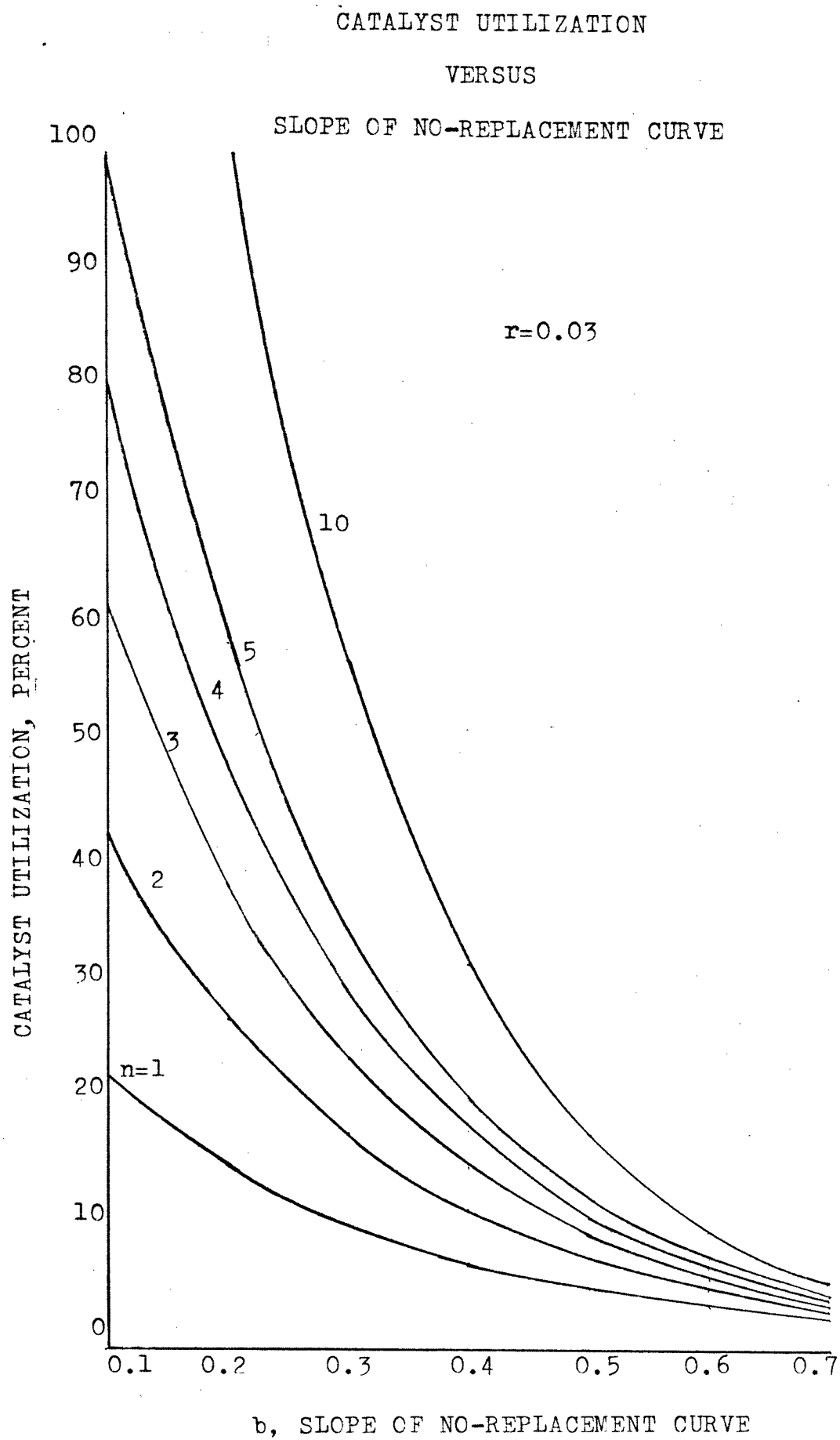


FIGURE 8

CATALYST UTILIZATION

VERSUS

SLOPE OF NO-REPLACEMENT CURVE

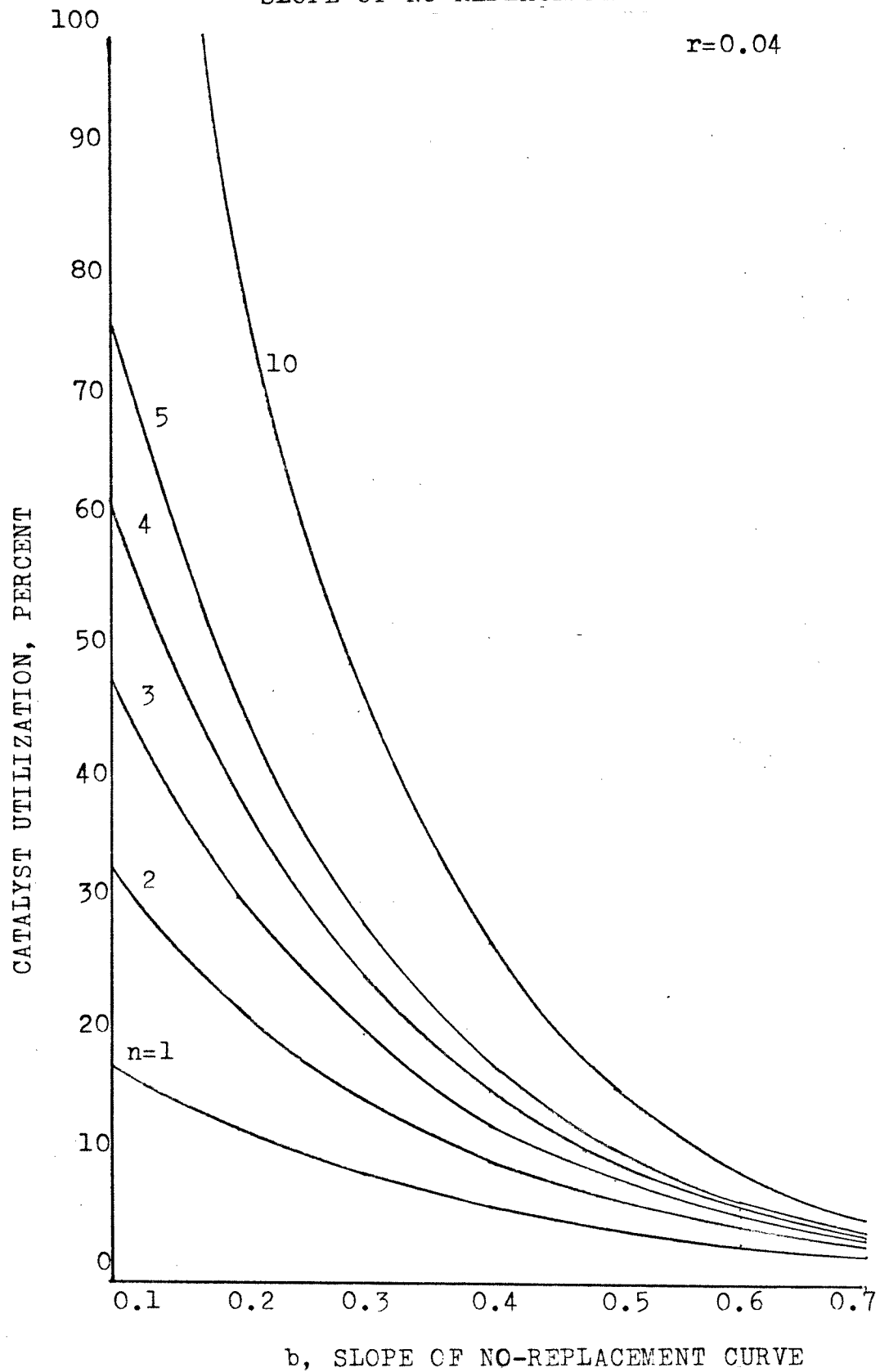


FIGURE 9
CATALYST UTILIZATION
VERSUS

SLOPE OF NO-REPLACEMENT CURVE

$r=0.05$

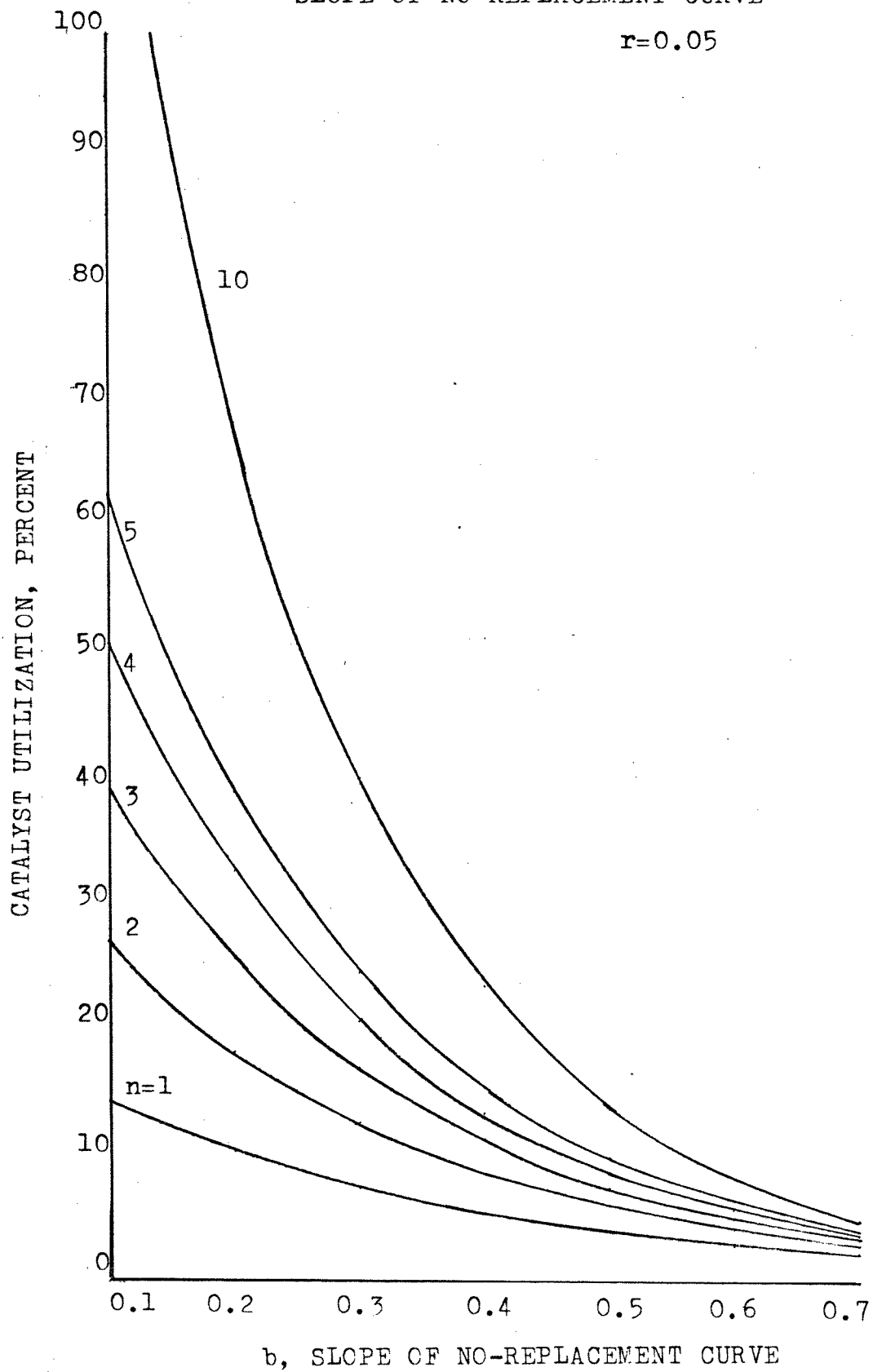


FIGURE 10

CATALYST UTILIZATION
VERSUS
SLOPE OF NO-REPLACEMENT CURVE

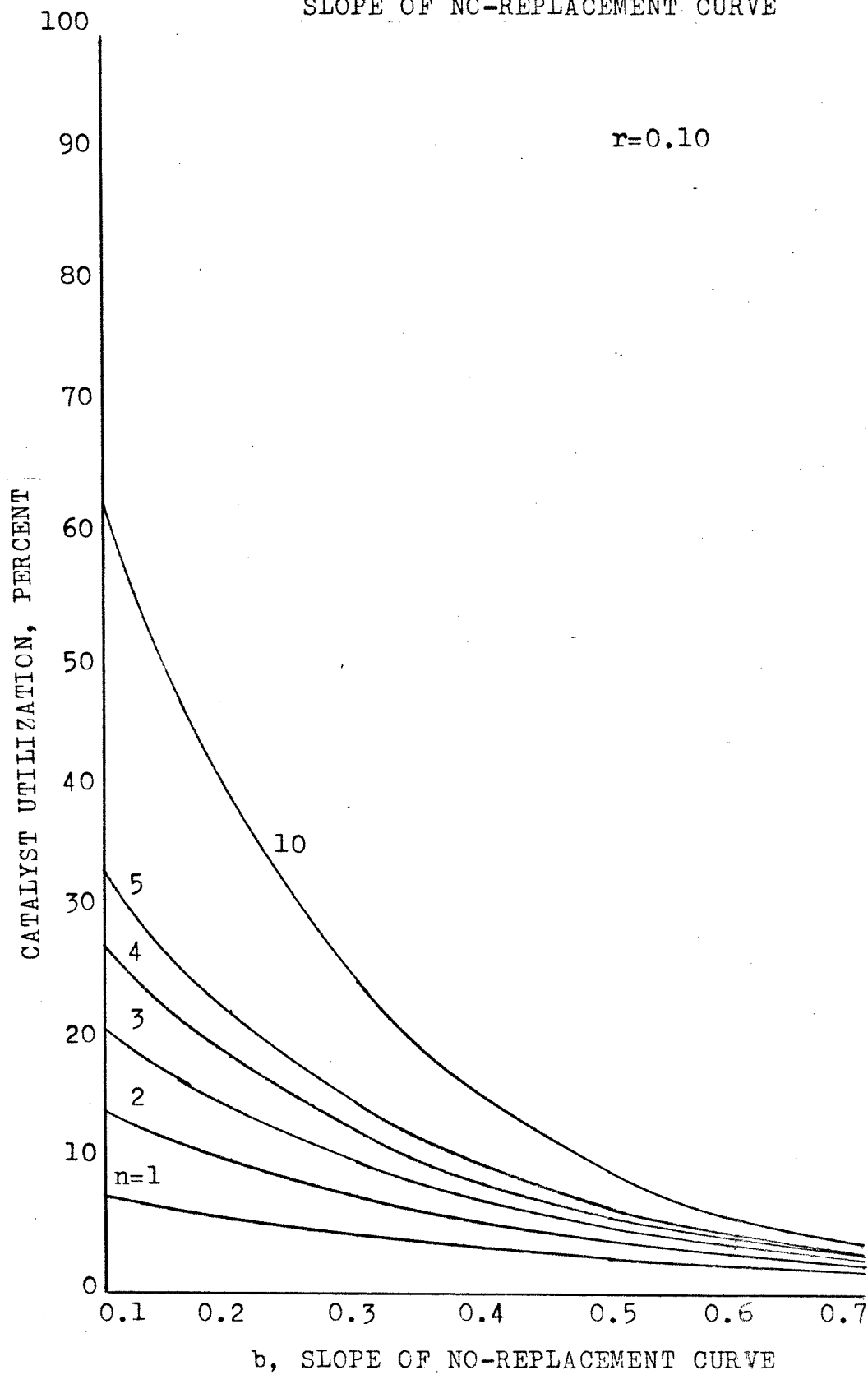
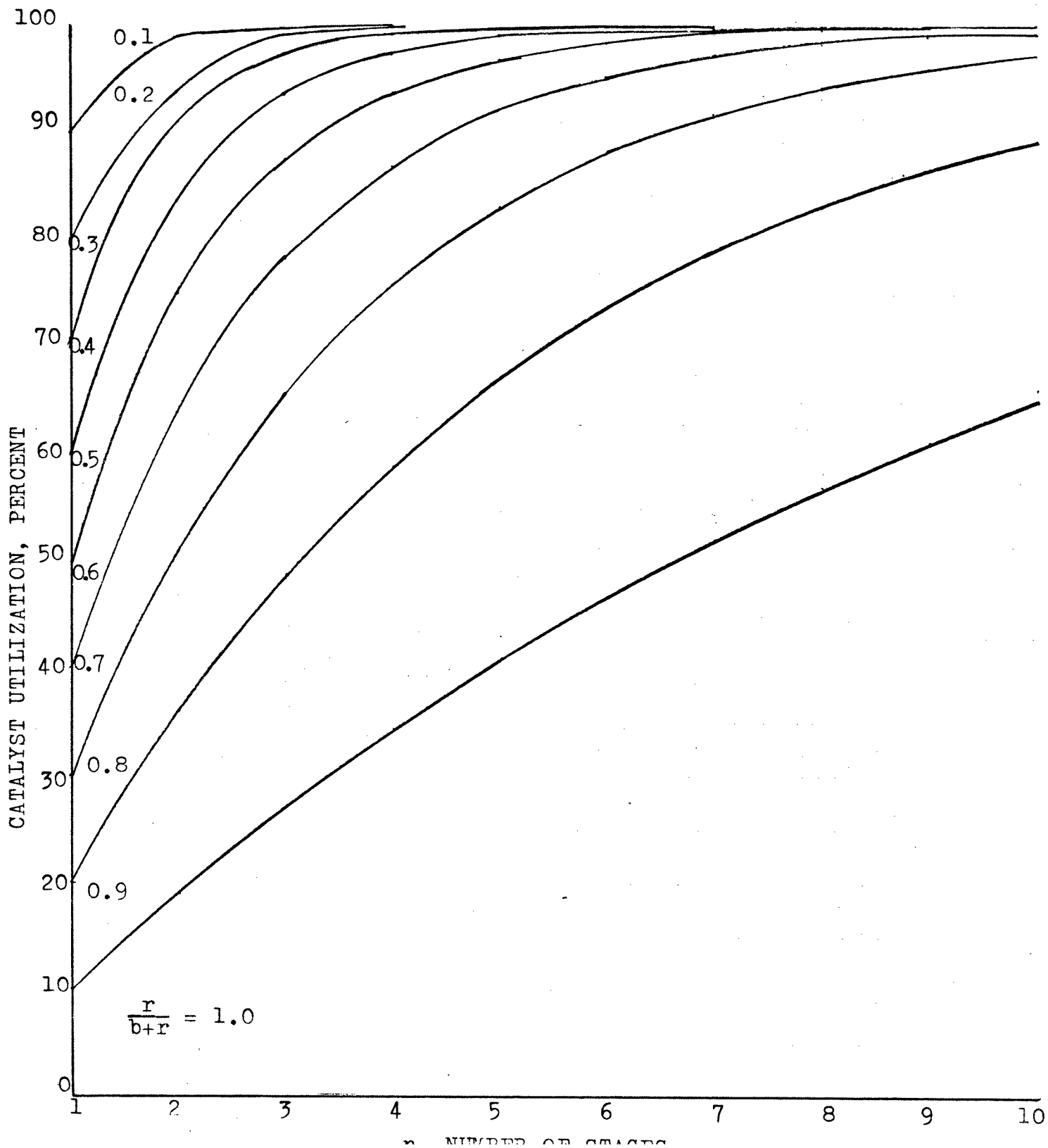


FIGURE 11

CATALYST UTILIZATION
 VERSUS
 NUMBER OF STAGES FOR SEMILOG DECLINE



Conclusions

With the use of Equation 11, one can then predict catalyst utilization for multistage fluidized reactors from laboratory catalyst activity data. The prediction equation is applicable for when activity decays as a log-log function, mixing is ideal, and constant inventory is maintained in each reactor.

The prediction equation for catalyst utilization enables one to make an economic study to determine the optimum balance among reactor costs, operating difficulties and catalyst utilization.

The prediction equation can promote savings in pilot plant and plant studies through elimination of repetitious experiments, since only one pilot plant run is needed to determine the slope at a particular set of conditions in a batch run with no replacement.

APPENDIX

TABLE 1

a, b	=	coefficient in catalyst-activity expression; $y=at^{-b}$
B1	=	calculates Gamma function in the Normgamm program for $n-b+1$ in Equation 11.
B2	=	calculates Gamma function in the Normgamm program for n in Equation 11.
C	=	catalyst utilization in the Normgamm program.
n	=	number of reactor in series.
p	=	fraction of catalyst which is age (probability density fraction for age of catalyst).
P	=	fraction of catalyst which is age t or less.
r	=	rate of replacement of catalyst, fraction per hour.
R_{I_u} , R'_{I_u}	=	catalyst utilization, based on ultimate no-replacement yield.

- t = time, hour.
- y = catalyst activity index, lb. product/
lb. catalyst/hr.
- Y_{r_1} = yield of product for continuous catalyst
replacement, one stage, lb. product/
lb. catalyst.
- Y_{r_n} = yield of product for continuous catalyst
replacement, n stages, lb. product/
lb. catalyst.
- Y_t = yield of product for no-replacement of
catalyst for catalyst of age t ,
lb. product/lb. catalyst.
- Y_u = ultimate yield for no-replacement of
catalyst, lb. product/lb. catalyst.

APPENDIX

TABLE 2

REFERENCES

Main

1. Anderson, S. C., and R. H. Matthias, Industrial Engineering Chemistry, vol. 46, no. 6, 1954, p. 1296.
2. Resnick, W., Industrial Engineering Chemistry, vol. 50, no. 10, 1960, p. 865.
3. MacMullin, R. B., and M. Weber, Transactions of American Institute of Chemical Engineers, vol. 31, 1935, p. 409.
4. Peirce, B. O., and R. H. Foster, A Short Table of Integrals, Fourth Edition, Blaisdell Publishing Company, New York, N. Y., p. 68.
5. Kreyszig, E., Advanced Engineering Mathematics, Second Edition, John Wiley & Sons, Inc., New York, N. Y., p. 711.

General

- Emmett, P. M., editor, Catalysis Fundamental Principles, vol. I, Reinhold Publishing Company, New York, N. Y., 1954, pp. 299-310.
- Emmett, P. M., editor, Catalysis Fundamental Principles, vol. III, Reinhold Publishing Company, New York, N. Y., pp. 152-164.
- Lee, W., Industrial Engineering Chemistry Process Design and Development, vol. 9, no. 1, 1970, p. 154.
- Pozzi, A. L., and H. F. Race, Industrial Engineering Chemistry, vol. 50, no. 7, 1958, p. 1075.
- Weekman, V. W., Jr., Industrial and Engineering Chemistry Process Design and Development, vol. 7, no. 1, 1968, p. 90.