APPENDIX G

PREDICTION OF RICHARDSON-ZAKI PARAMETERS FOR PDU EXPERIMENTAL MODELING

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BED EXPANSION OF CYLINDRICAL CATALYSTS IN LIQUID FLUIDIZATION

ABSTRACT

Fluidization of cylindrical particles by pure fluids and by coal slurries has been studied. It is shown that bed expansion data of cylindrical-shaped particles in liquid fluidization are adequately correlated using the Richardson-Zaki method. Particle terminal velocities are shown to be well predicted using the drag coefficient--Reynolds number relationship for infinte cylinders, with the characteristic dimension given by the cylinder diameter. The original correlations suggested by Richardson and Zaki for their index, n, in terms of particle Reynolds number are shown to be satisfactory.

INTRODUCTION

Nearly all gas-solid and liquid-solid fluidization data have been obtained in beds of spherical or near-spherical particles. Fluidization data for cylindrically shaped pellets are scarce. Table I lists only four sources where these kinds of data are available. In view of the recent interest in the catalytic hydroliquefaction of coal (the H-Coal process) and the hydroprocessing of petroleum resids, a better understand of the fundamentals of ebullated (or three-phase fluidized) beds is essential for successful scale-up. The commercial catalysts used in these processes are nearly all in the form of cylindrical extrudates. The prediction of the hydrodynamic behavior of a three-phase fluidized bed from basic fluid and particle mechanics is not possible at present. This applied even to basic parameters such as bed expansion and phase holdup. At least two phenomenological models have been proposed: namely, those of Bhatia and Epstein (1), and

Darton and Harrison (5). However, their correlations for gas holdups were borrowed entirely from the two-phase literature, and a more definitive analysis seems necessary. As for the treatment of the particulate phase (i.e., phase excluding the bubble and its associated wake), both models suggest the use of the Richardson-Zaki correlation (8). Thus it is crucial to determine how well the Richardson-Zaki correlation can predict liquid fluidized bed behavior of cylindrical particles. For such a study, Vasalos et al (12) found that the data could be correlated by the Richardson-Zaki type of correlation, but the two parameters (i.e., terminal velocity and index n) in the Richardson-Zaki correlation were fit empirically using the data. The present study is an extension of Vasalos' work. In addition, methods are suggested for determining the two parameters in the Richardson-Zaki correlation.

DISCUSSION

The Richardson-Zaki correlation (8) has been widely used to correlate gas-solid and liquid-solid fluidization data. Basically, the model relates the liquid holdup, ϵ_1, to the sperficial liquid velocity, j_1, and the terminal velocity of the solid, U_t:

$$\epsilon_1^n = j_1/U_t \qquad (1)$$

where n is the so-called Richardson-Zaki index. Richardson and Zaki used the drag-coefficient relationship to estimate U_t and proposed to correlate n as a function of particle Reynolds number.

Usually the settled bed porosity, ϵ_{1o}, and bed height, H_o, are known, and the material balance on the solids yields:

$$(1 - \epsilon_1)H = (1 - \epsilon_{1o})H_o \qquad (2)$$

Thus, bed expansion can be predicted once the liquid holdup inside the bed is found using Equation 1.

Terminal Velocity of a Cylindrical Particle

The terminal velocity of a particle falling in a fluid is determined by balancing the gravitational and drag forces. For spherical particles of diameter, d, we have:

$$\pi d^3 g\ (\rho_s - \rho_1)/6 = C_D(\pi d^2/4)(\rho_1 U_t^2/2) \qquad (3)$$

where the nomenclature has the usual meaning. The drag coefficient, C_D, is a function of the particle Reynolds number, Re_t, defined by:

$$Re_t = \rho_1 U_t d/\mu_1 \qquad (4)$$

For example, the drag coefficients for spheres, disks, and cylinders can be found in Perry (10) and are shown in Figure 1. In order to calculate the terminal velocity, one has to resort to trial and error using Figure 1. However, one can simplify Equation 3 and replace U_t with Re_t, thus obtaining:

$$C_D Re_t^2 = 4/3 \left[d^3 \rho_1 g(\rho_s - \rho_1)/\mu_1^2 \right] = (4/3)Ga, \text{ for spheres} \quad (5)$$

where Ga is the Galileo number and can be evaluated solely from known quantities. Since C_D is a function of Re_t, $C_D Re_t^2$ can be expressed analytically or graphically as a function of Re_t. The resulting graphical relationship based on Figure 1 is shown in Figure 2 for all three shapes, and therefore the Reynolds number, Re_t, and hence terminal velocity, U_t, can be derived.

Terminal velocity measurements were made by dropping single HDS-2A (American Cyanamid) catalyst extrudates in various fluids. Since the catalyst is porous in nature, it was thoroughly soaked in the particular fluid studied before testing. The soaked catalyst density was then determined by simple volume displacement and weighing methods. The results are summarized in Table II. Experimentally, the catalyst was observed to fall with its axis horizontal, even in cases where it was released from the vertical position. The following methods were used to correlate the data.

Method A--The pellet diameter, d, was used as the characteristic length. Here, a similar relationship can be derived between $C_D Re_t^2$ and the Galileo number; namely:

$$C_D Re_t^2 = (\pi/2)\,Ga, \text{ for cylinders} \quad (6)$$

The experimental data are plotted in Figure 3, where the data exhibited good agreement with the infinite cylinder C_D vs. Re_t relationship.

Method B--For this calculation of the dimensionless groups (Re_t, Ga), the equivalent particle diameter, d_s, (defined as the diameter of a sphere having the same volume) is used. For a cylindrical pellet:

$$d_s = (3d^2 \ell/2)^{1/3} \quad (7)$$

The results shown in Figure 4 indicate that the data lie significantly below the sphere relationship; hence, it is concluded that d_s is not a suitable dimension to be used. Note that this is the characteristic diameter used by Vasalos et al (12) and their proposed correlation is also shown in Figure 4 for comparison. It is not surprising that the Vasalos et al correlation fit the data in this form quite well because d_s was used to calculate these data points.

Method C--Heywood (7) suggested using the projected diameter, d_h, equal to the diameter of the circle, having an area equal to the projected area of the particle when placed in the most stable position. In the present case, d_h is given by:

$$d_h = (4d\ell/\pi)^{1/2} \quad (8)$$

The terminal velocity data reduced by Heywood's approach are presented in Figure 5. The agreement with the sphere C_D vs Re_t relation is poor.

Method D--In packed column studies, the usual mean particle diameter, d_m, (see Bird, et al, 1960) is given by:

$$d_m = 6/a_v \quad (9)$$

where the specific surface, a_v, is the ratio of particle surface area to particle volume. It can easily be shown that d_m is related to d_s via sphericity, ϕ_s; i.e.:

$$d_m = \phi_s d_s \quad (10)$$

where the sphericity is defined as:

ϕ_s = (surface area of sphere/surface area of particle, both of the same volume)

and hence for a cylindrical-shaped particle

$$\phi_s = (3/2\ d^2 \ell)^{2/3} / \left[(d^2/2) + d\ell\right] \quad (11)$$

The terminal velocity data using d_m as the characteristic length are shown in Figure 6. As a whole, the drag relationship for a sphere correlates the data very well at Reynolds numbers less than 100, but tends to overestimate the terminal velocity at higher Reynolds numbers.

Among the four methods tried, the best correlation is obtained by using the pellet diameter as the characteristic length in conjunction with the C_D vs Re relationship for infinite cylinders. This analysis demonstrates that the drag coefficient for finite cylinders of ℓ/d equal to 3 approaches closely to that for infinite cylinders.

Bed Expansion

According to the Richardson-Zaki correlation, bed expansion data can be reduced on a plot of log j_1 versus log ϵ_1. A straight line can be drawn through the data:

$$\log j_1 = \log U_t + n \log \epsilon_1 \quad (12)$$

The intercept at $\epsilon_1 = 1$ yields log U_t and the slope of the line gives the value for n. All the available data were reduced in this fashion. Liquid holdup was evaluated based on the expanded bed height. In the case of H-Coal AU-77H data (11), the kerosene/coal fines slurry holdup was obtained from bed height measurements using a travelling gamma-ray densitometer. All of the results are summarized in Table III, and the corresponding Richardson-Zaki plots can be found in Appendix A. As a whole, the data were found to be well correlated by a straight line on these plots, thus confirming the validity of the form of the correlation proposed by Richardson and Zaki.

Viscosity

For pure liquids such as kerosene and mineral oil, the liquid viscosity, μ_o, can be easily measured or obtained from physical property tables. However, for the kerosene/coal fines slurry employed in the H-Coal modeling study, the mixture (effective) viscosity, μ_m, is dependent on the solid content. In general, it was found (6) that the relative viscosity (μ_m/μ_o) is a function of the solid volume fraction. The mixture viscosity data (11) for kerosene slurry were so reduced, as shown in Figure 7, and a best linear fit was found:

$$\mu_m = \mu_o\,(1 + 26.8\,\epsilon_s) \qquad (13)$$

The dimensionless numbers in Table III for the kerosene/coal fines slurry fluidization data were all evaluated using Equation 13 for the viscosity values.

Terminal Velocity

The terminal velocity data extrapolated using the Richardson-Zaki method were plotted as terminal velocity Reynolds number versus the Galileo number. Figure 8 shows that on a plot of Re_t versus Ga using d as the characteristic length, the terminal velocity data can be well correlated by the drag coefficient relationship for infinite cylinders. However, the mineral oil data from the H-Coal AU-77H unit were found to lie significantly lower than other data in Figure 8. One possible explanation is that the catalyst was not completely soaked before the mineral oil experiments were conducted. Table IV compares the predicted terminal velocities with the experimental terminal velocities from Richardson-Zaki plots. The overall mean ratio of predicted value to observed value is 0.967, although the mean ratio for mineral oil data is only 0.737. This analysis demonstrates that the terminal velocities for cylindrical pellets in a fluidized bed can be predicted using the C_D vs Re_t relationship for infinite cylinders with the characteristic dimension given by the pellet diameter.

Correlation of Index, n

Richardson and Zaki found the index, n, to be a function of the particle Reynolds number as follows:

$$\begin{array}{ll} Re < 0.2 & n = 4.65 \\ 0.2 < Re < 1 & n = 4.35\,Re^{-0.03} \\ 1 < Re < 500 & n = 4.45\,Re^{-0.1} \\ 500 < Re & n = 2.39 \end{array} \qquad (14)$$

However, this relationship was obtained for spherical particles only. Values of n for the present data are plotted in Figure 9 as a function of Galileo number. Since a unique relationship exists between Reynolds number and Galileo number for spherical particles, Equation 14 can easily be transformed to yield n as a function of Ga. The resulting curve as shown in Figure 9 is seen to slightly overpredict the value of n. Values of n are also presented as a function of particle Reynolds number as shown in Figure 10. The original Richardson-Zaki correlation for n (Equation 14) is seen to correlate the data slightly better in terms of Reynolds number. However, more data are needed to confirm the validity of this correlation for cylindrical particles, particularly in the laminar ($Re < 1$) and turbulent ($Re > 1000$) regimes.

Richardson and Jeronimo (9) have also recently investigated methods of estimating n. One method that appears quite promising is to assume Equation 1 to be valid at both the minimum fluidization point and the fully expanded bed of voidage unity. The minimum fluidization velocity, U_{mf}, was estimated using the Ergun equation (2) for packed beds. The value of n is hence given by:

$$n = \frac{\log U_{mf} / U_t}{\log \epsilon_{mf}} \tag{15}$$

where ϵ_{mf} is the voidage at minimum fluidization. Their results indicate that values of n calculated using Equation 15 and the Ergun equation are considerably higher than the original Richardson-Zaki values, i.e., Equation 14. Richardson and Jeronimo attributed the discrepancy to the validity of using Ergun's equation for a fluidized bed and the uncertainty of choosing the value for ϵ_{mf}. We feel that the Richardson-Zaki correlation (Equation 1) does not apply near the minimum fluidization point and the original experimental data of Richardson and Zaki tend to support this observation. Richardson and Jeronimo's data also show that the Richardson-Zaki approach does not apply at the minimum fluidization point, which resulted in the larger values of n being obtained. Thus, Equation 15 is not recommended as a method for evaluation of n. Equation 14 is the best available at the present time.

CONCLUSIONS

1) The Richardson-Zaki type correlation can be used to analyze bed expansion data of cylindrical-shaped particles in both pure liquid and coal slurry fluidization.

2) The terminal velocity values obtained from both single cylindrical catalyst drop experiments and liquid fluidized bed experiments can be adequately correlated using the C_D versus Re_t relationship for infinite cylinders with the characteristic dimension given by the pellet diameter. Three other characteristic dimensions were tried in attempting to correlate the single pellet terminal velocity data, but none was found to be satisfactory.

3) The Richardson-Zaki index, n, obtained from data used in this study is presented both as a function of the Galileo number and the Reynolds number. The original Richardson-Zaki correlation for index n is found to correlate present data better in terms of Reynolds number. More data are required, however, to confirm the validity of this correlation at both low and high Reynolds number.

REFERENCES FOR APPENDIX G

G-1) Bhatia, V. K., and N. Epstein, "Three-Phase Fluidization: A Generalized Wake Model," Proc Euro Symp on Fluidization, 380 (1974).

G-2) Bird, R. B., et al., Transport Phenomena, John Wiley and Sons (New York) (1960).

G-3) Chemical Systems, Incorporated, "Liquid Phase Methanation," R&D NPT No. 78, Interim Report No. 1 (1973).

G-4) Darton, R. C., and D. Harrison, "Gas and Liquid Holdup in Three-Phase Fluidization," Chem Eng Sci, 30, 581 (1975).

G-5) Happel, J., and H. Brenner, Low Reynolds Number Hydrodynamics with Special Applications to Particulate Media, Chapter 9, Prentice-Hall (New Jersey) (1965).

G-6) Heywood, H., "Uniform and Non-uniform Motion of Particles in Fluids," Sym Interaction Between Fluids and Particles, Inst Chem Eng (London) (1962).

G-7) Richardson, J. F., and W. N. Zaki, "Sedimentation and Fluidization: Part I," Trans Inst Chem Eng, 32, 35 (1954).

G-8) Richardson, J. F., and M. Jeronimo, "Velocity-Voidage Relations for Sedimentation and Fluidization," Chem Eng Sci, 34, 1419 (1979).

G-9) Perry, J. H., Chemical Engineers' Handbook, Fourth Edition, McGraw-Hill (New York) (1963).

G-10) Vasalos, I. A., et al., "Study of Ebullated Bed Fluid Dynamics for H-Coal," Final Report, M80-21 (1980).

G-11) Vasalos, I. A., et al., "Holdup Correlations in Slurry Solid Fluidized Beds," AIChE J, 28, 346 (1982).

G-12) Wolk, R., "Gas-Liquid-Solid Fluidization," MS Thesis, Polytechnic Institute of Brooklyn (1962).

TABLE I

LIQUID FLUIDIZATION DATA FOR CYLINDRICAL CATALYSTS

Source	Catalyst	Fluid	Test Column Diameter (cm)
Amoco Oil (Vasalos, 1980)	HDS-2A d = 0.16 cm l = 0.51 cm	Kerosene/Coal Fines Slurry Mineral Oil (Amoco 9 NF)	15.2
HRI (Wolk, 1962)	Nalcomo 471 d = 0.079 cm l = 0.48 cm	2-Propanol	2.5
Chem Systems, Inc. (1973)	Methanation Catalyst d = 0.16 cm l = 0.32 cm	Hexane	2.2

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TABLE II

SUMMARY OF TERMINAL VELOCITY MEASUREMENTS OF INDIVIDUAL CYLINDRICAL PELLET

Pellet	Particle Diameter, cm	Particle Length, cm (1)	Fluid	Temp, ^oC	Fluid Density g/cm^3	Viscosity, cP	Soaked Density, g/cm^3	Measured (2) Terminal Velocity, cm/s
HDS-2A	0.16	0.5	Water	20	1.0	1.0	1.79	14.4
HDS-2A	0.16	0.5	Mineral Oil	66	0.82	6.2	1.61	11.9
HDS-2A	0.16	0.5	Mineral Oil	79	0.81	4.2	1.61	13.0
HDS-2A	0.16	0.5	Kerosene	20	0.79	1.39	1.68	16.4,17.0
HDS-2A	0.16	0.5	Ethylene Glycol	20	1.11	19.9	1.76	6.0
Glass Rod	0.20	0.5	Ethylene Glycol	20	1.11	19.9	2.23	10.8

(1) All Lengths are nominal values.

(2) At least 20 particles were dropped and the velocities reported are statistical averages.

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TABLE III

TABULATION OF RICHARDSON-ZAKI PARAMETERS FROM LIQUID FLUIDIZATION DATA

Source	Catalyst	Extrudate Diameter (cm)	l/d	Run No.	Fluid	Coal Fines Conc (Vol%)	Temp °C	Soaked ρ(Cat.) g/cm^3	ρ(Slry) g/cm^3	Viscosity (cP)	U_t (cm/s)	Index n	Re_t	Ga
Amoco Oil (Vasalos, 1980)	HDS-2A	0.16	3.2	201(1)	Kerosene	0	2[illegible]	1.64	0.79	1.39	14.0	2.66	127	13,970
				301	Kerosene	0	22	1.64	0.79	1.39	15.0	2.64	136	13,970
				204	Kerosene	5.1	21	1.64	0.84	3.34	13.8	3.0	55	2,420
				205	Kerosene	10.4	19	1.64	0.89	5.28	12.5	3.0	34	960
				210	Kerosene	11.9	22	1.64	0.90	5.84	10.7	2.89	26	790
				209	Kerosene	15.5	36	1.64	0.93	6.40	10.2	3.05	24	650
				208, 214	Kerosene	15.5	66	1.63	0.91	4.58	11.0	2.81	35	1,250
				420, 427	Mineral Oil	0	37	1.68	0.85	14.6	7.1	3.58	6.6	130
				421, 428	Mineral Oil	0	52	1.67	0.83	9.5	8.2	3.28	12	310
				422, 426	Mineral Oil	0	6[illegible]	1.66	0.82	6.0	9.0	2.94	20	800
				423, 429	Mineral Oil	0	79	1.66	0.81	4.2	10.3	2.83	32	1,570
Chem Sys, Inc.	Standard Methanation Catalyst	0.16	2.0	46-1	Hexane	0	20	2.25	0.66	0.3	25.0	2.6	868	3.67×10^5
HRI (Wolk, 1962)	Nalcomo 471	0.079	6.0	--	2-Propanol	0	20	1.42	0.79	2.5	9.6	2.81	24	391

(1) Run number corresponding to original report.

(2) This run was conducted in a 15.2 cm ID column.

JCL/ml
8/28/81

TABLE IV

TERMINAL VELOCITY PREDICTIONS FOR BED EXPANSION DATA

Source	Fluid	Run ID	Ga	Expt U_t (cm/s)	Pred U_t (cm/s)	Pred U_t/ Expt U_t	
Amoco Oil	Kerosene	201	13,970	14.0	15.3	0.915	
(Vasalos)	Slurry	301	13,970	15.0	15.3	0.980	
		204	2,420	13.8	13.4	1.030	
		205	960	12.5	12.4	1.008	$\bar{x}$ = 0.966 (1)
		210	790	10.7	11.4	0.939	σ = 0.041
		209	650	10.2	10.8	0.944	
		208,214	1,250	11.0	11.6	0.948	
	Mineral Oil	420,427	130	7.1	9.5	0.747	
		421,428	310	8.2	11.1	0.739	$\bar{x}$ = 0.737
		422,426	800	9.0	12.8	0.703	σ = 0.024
		423,429	1,570	10.3	13.6	0.757	
Chem Sys	Hexane	46-1	3.67×10^5	25.0	24.8	1.008	
HRI (Wolk)	n-Propanol	--	391	9.6	7.2	1.333	

(1) x = mean; = standard deviation.

JCL/ml
9/28/81

Figure 1

DRAG COEFFICIENTS FOR SPHERES, DISKS AND CYLINDERS (from Perry (10))

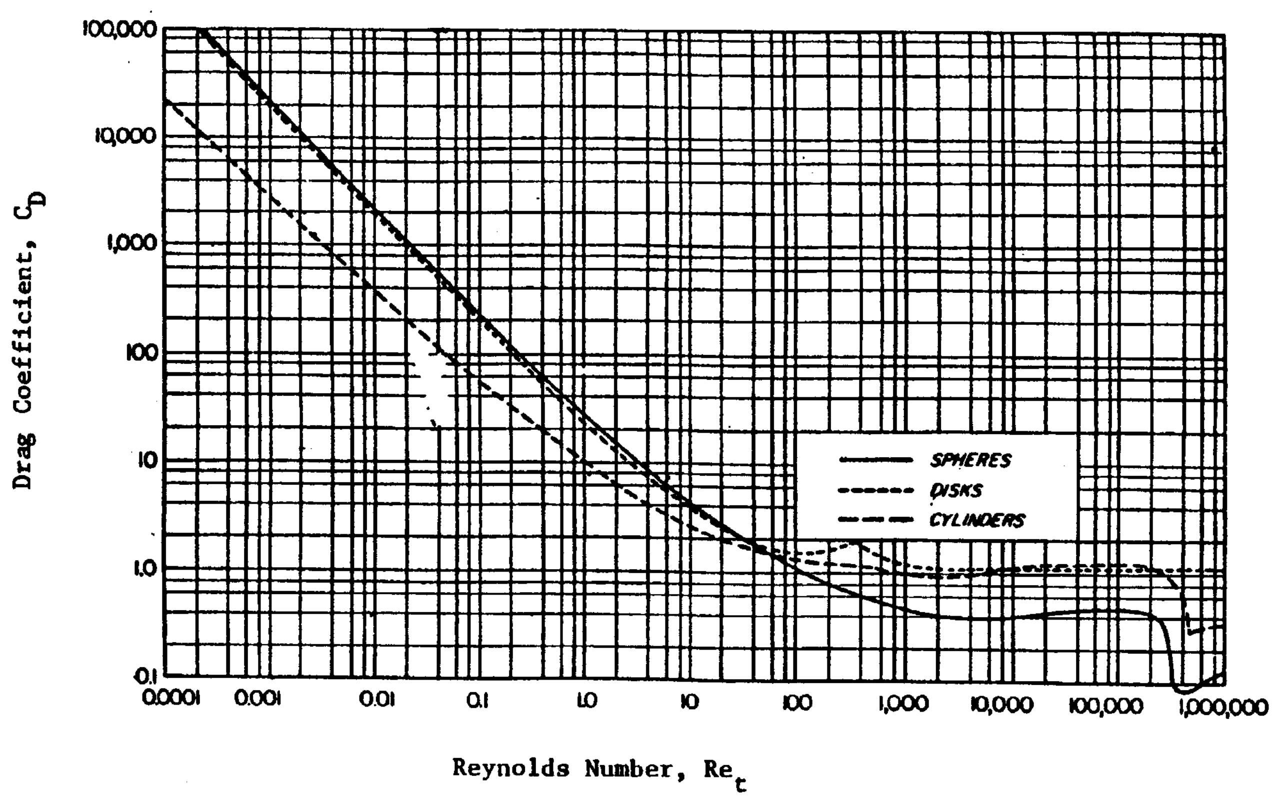
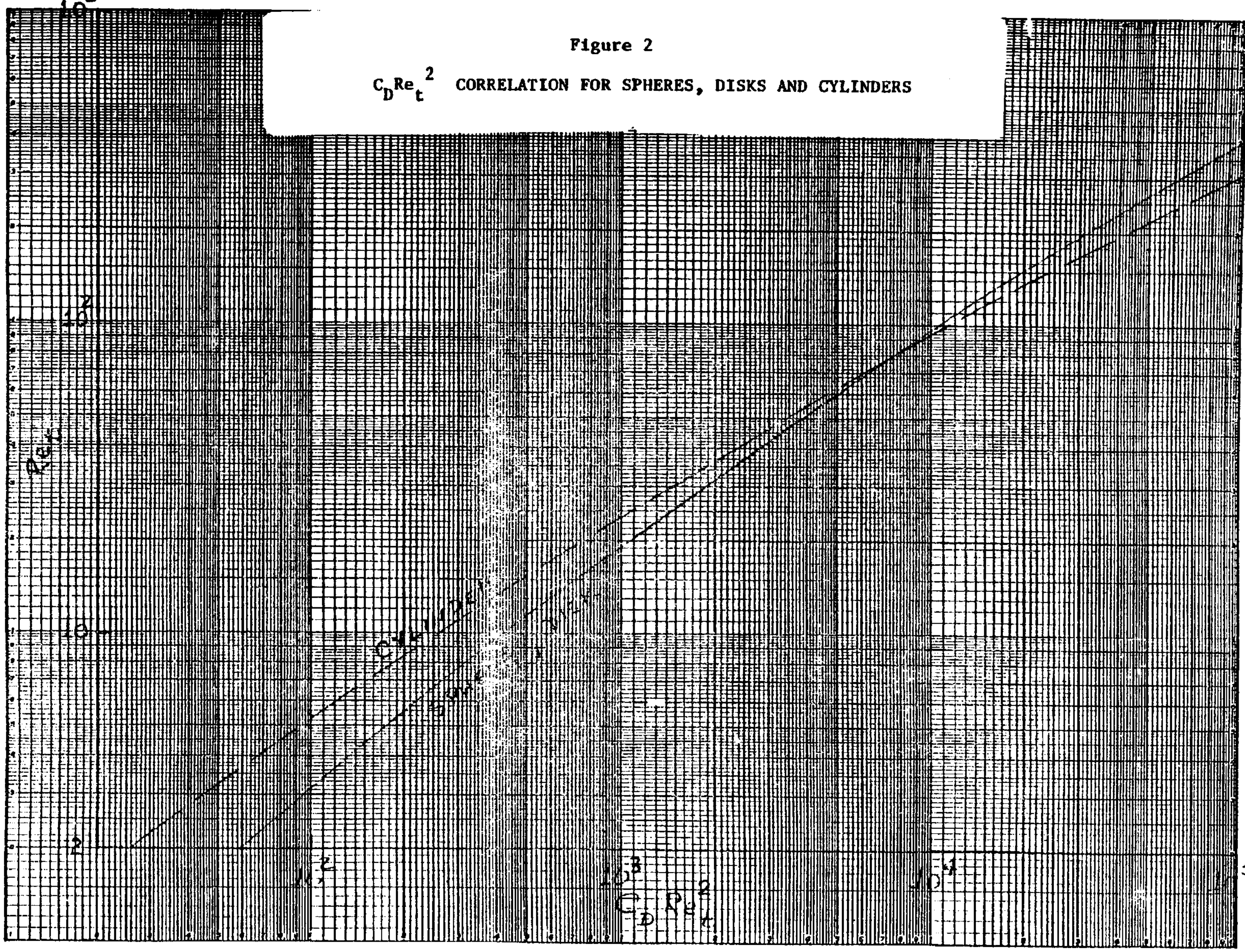


Figure 2

$C_D Re_t^2$ CORRELATION FOR SPHERES, DISKS AND CYLINDERS



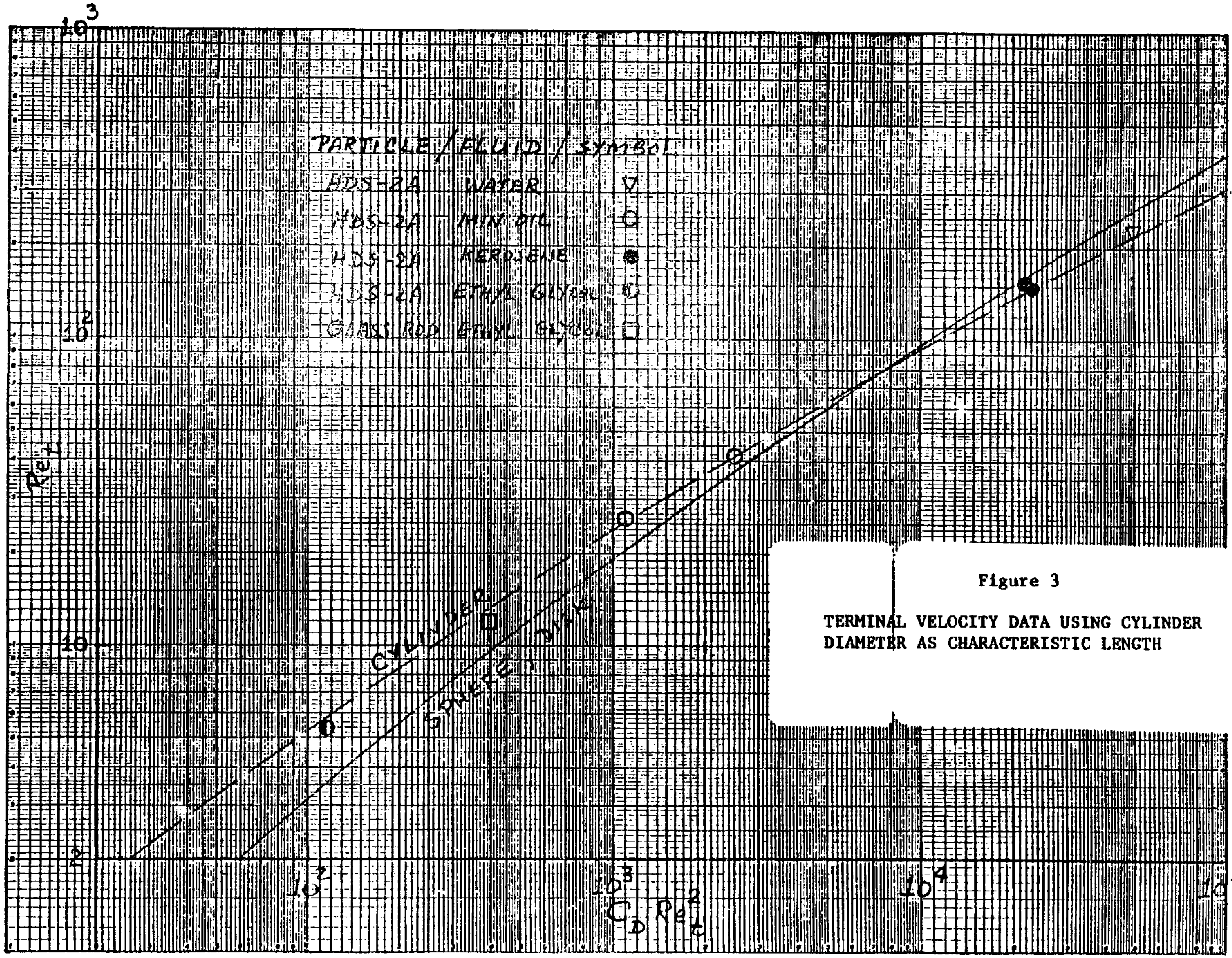


Figure 3

TERMINAL VELOCITY DATA USING CYLINDER DIAMETER AS CHARACTERISTIC LENGTH

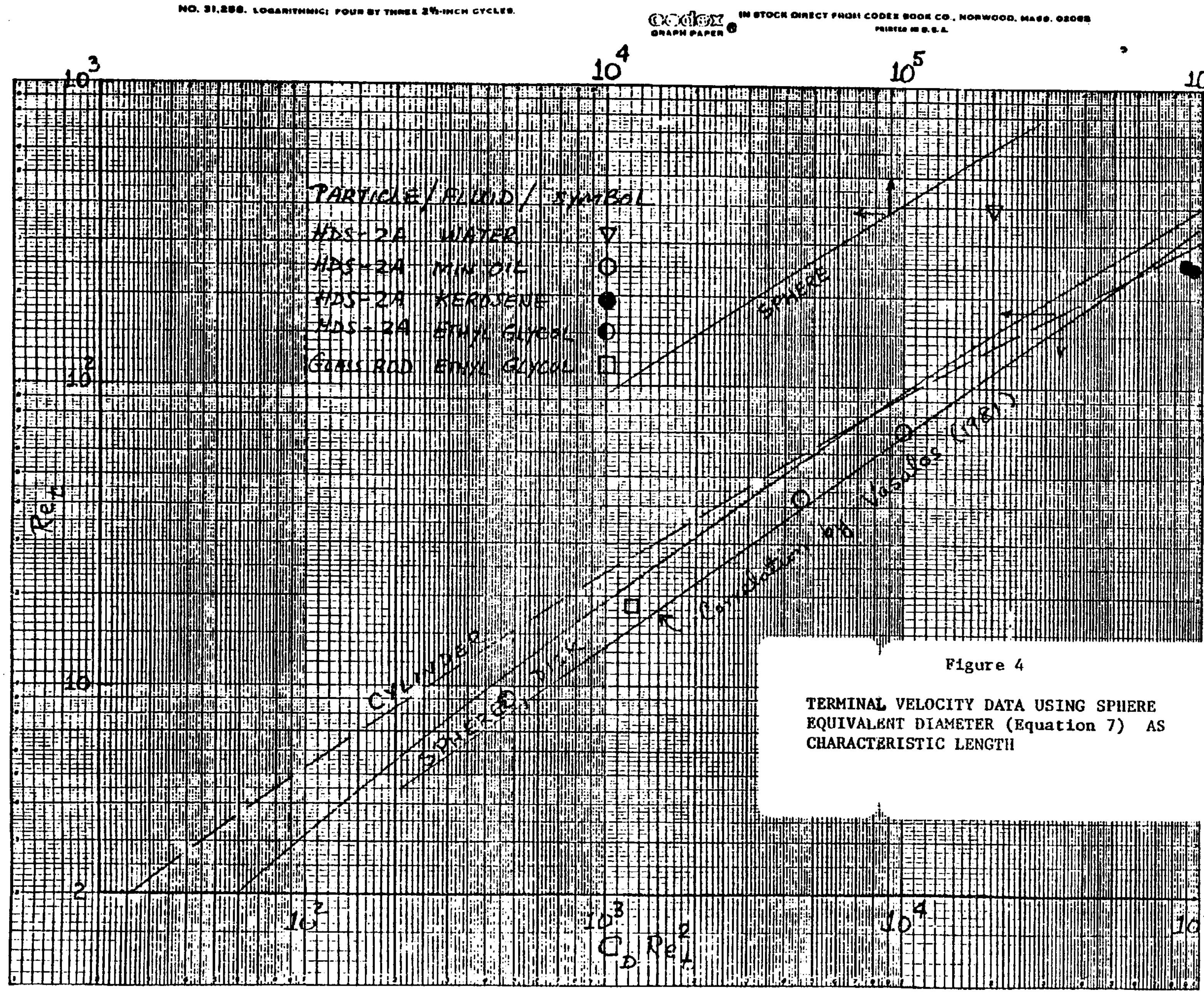


Figure 4

TERMINAL VELOCITY DATA USING SPHERE EQUIVALENT DIAMETER (Equation 7) AS CHARACTERISTIC LENGTH

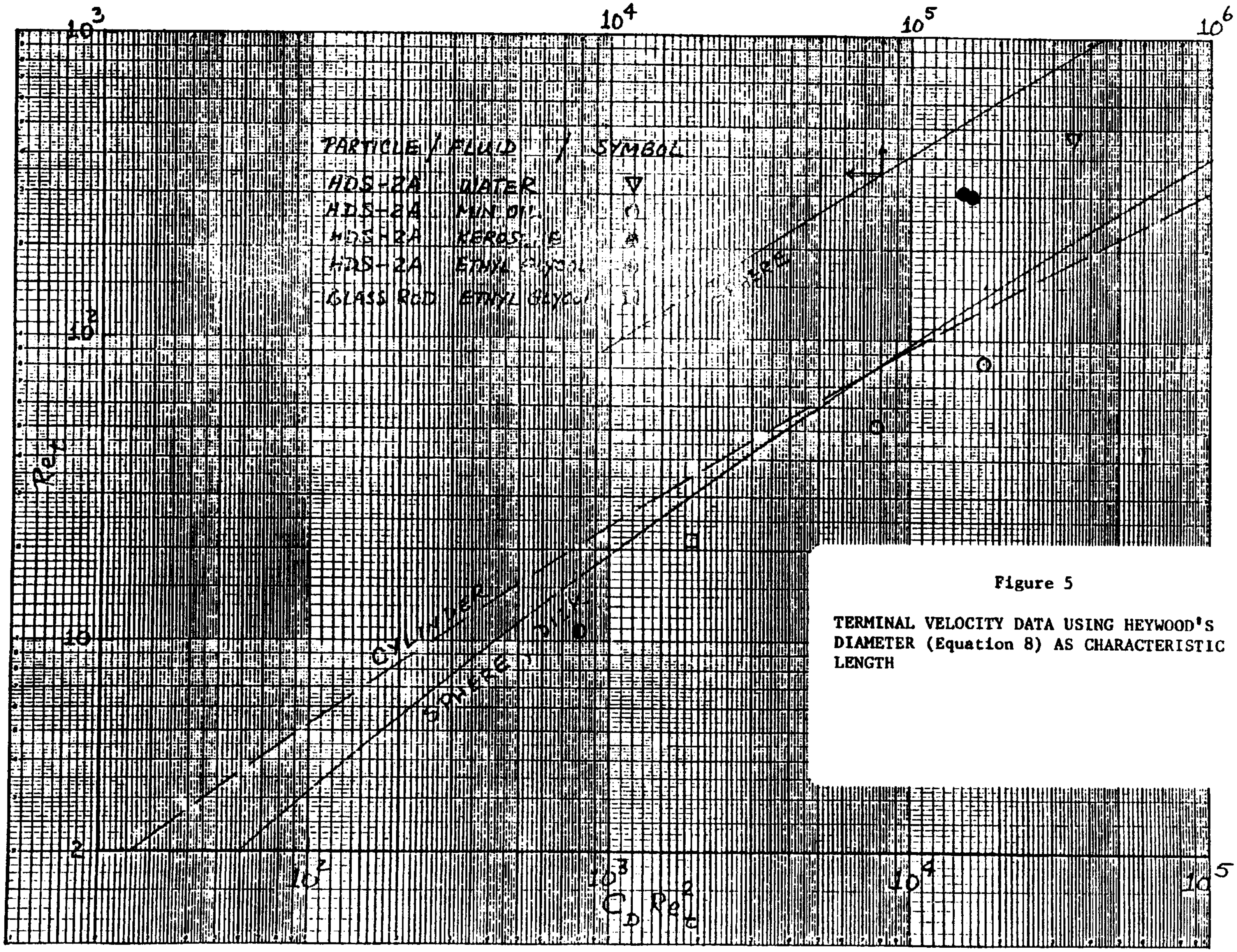


Figure 5

TERMINAL VELOCITY DATA USING HEYWOOD'S DIAMETER (Equation 8) AS CHARACTERISTIC LENGTH

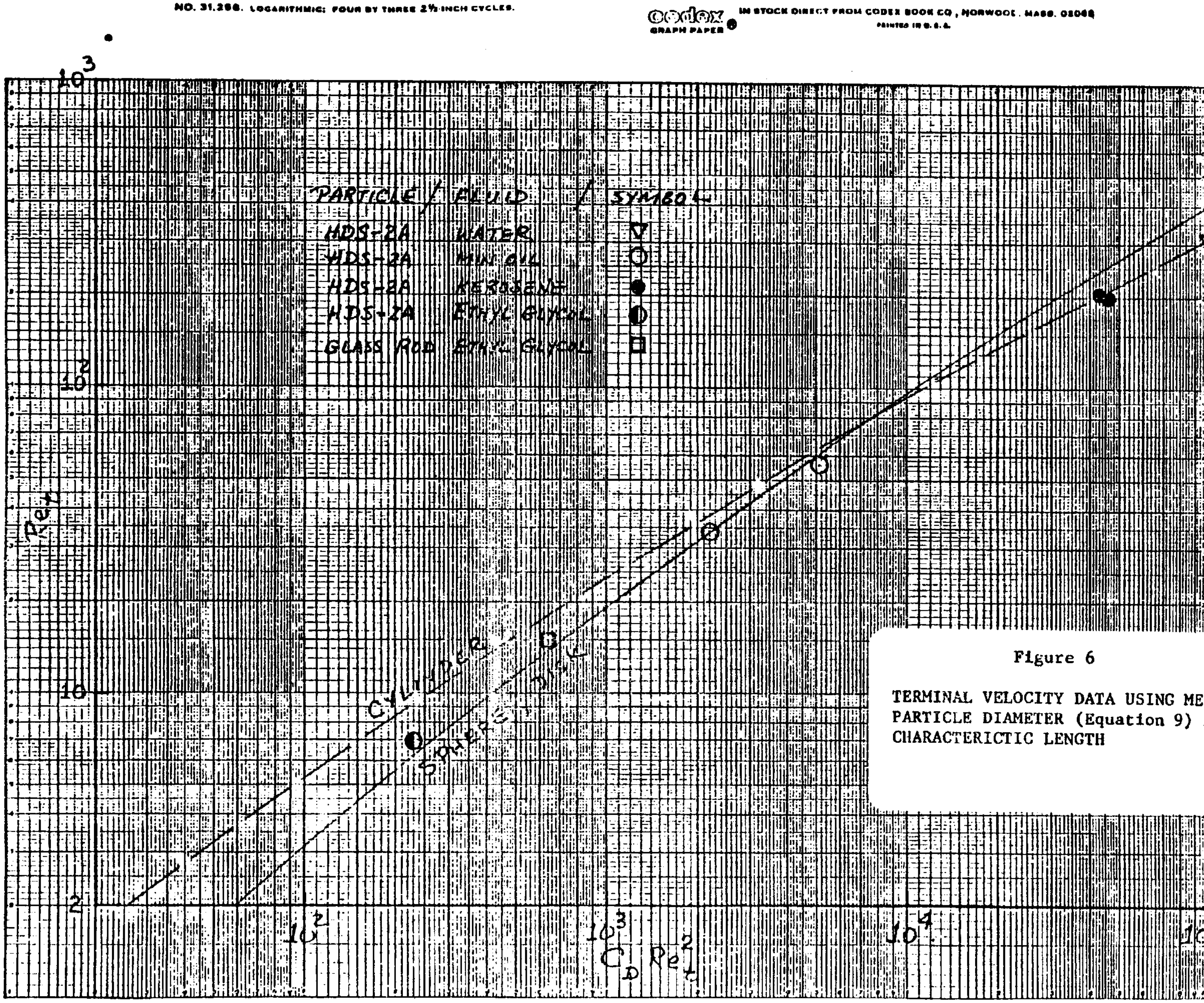
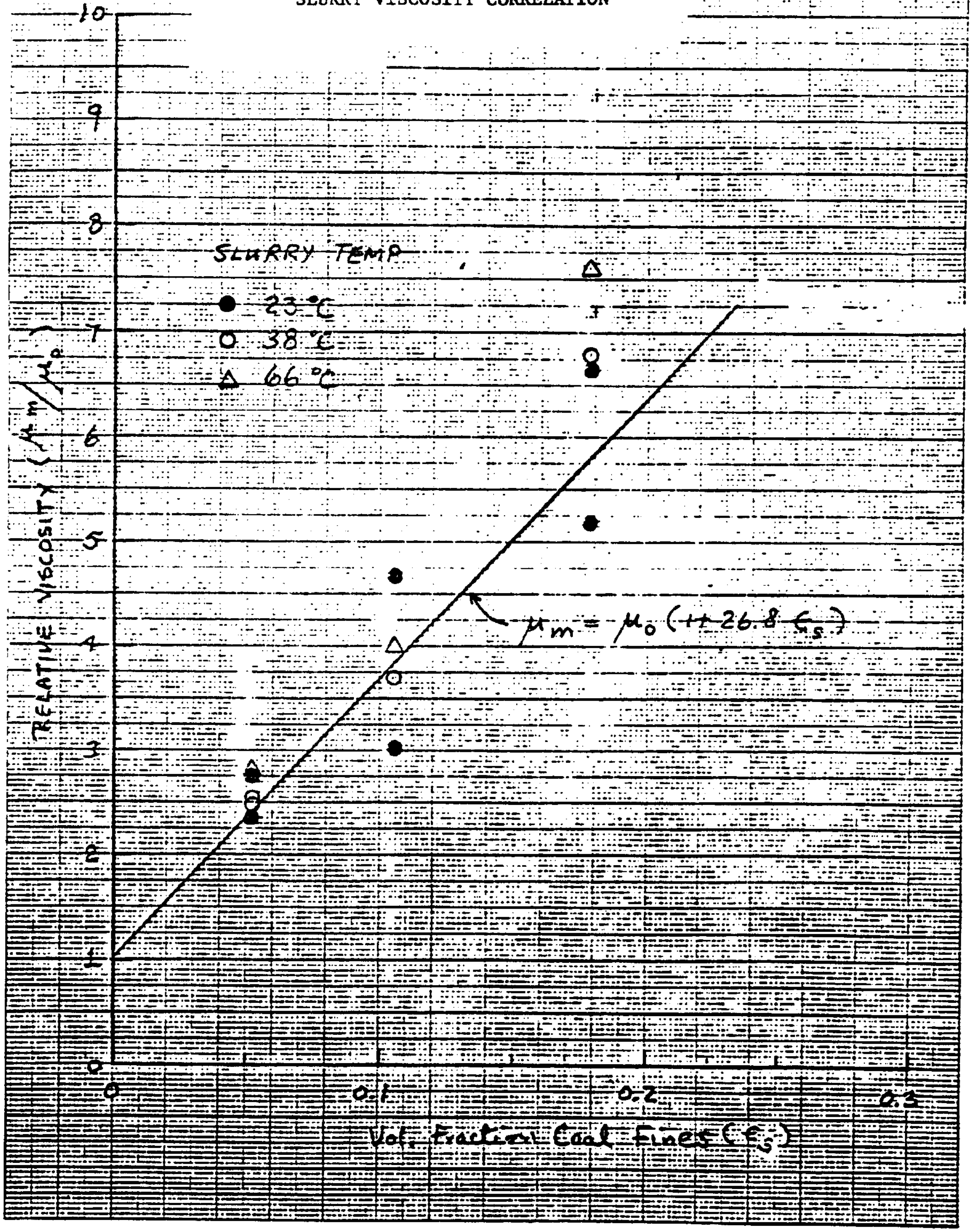


Figure 6

TERMINAL VELOCITY DATA USING MEAN PARTICLE DIAMETER (Equation 9) AS CHARACTERICTIC LENGTH

Figure 7

SLURRY VISCOSITY CORRELATION



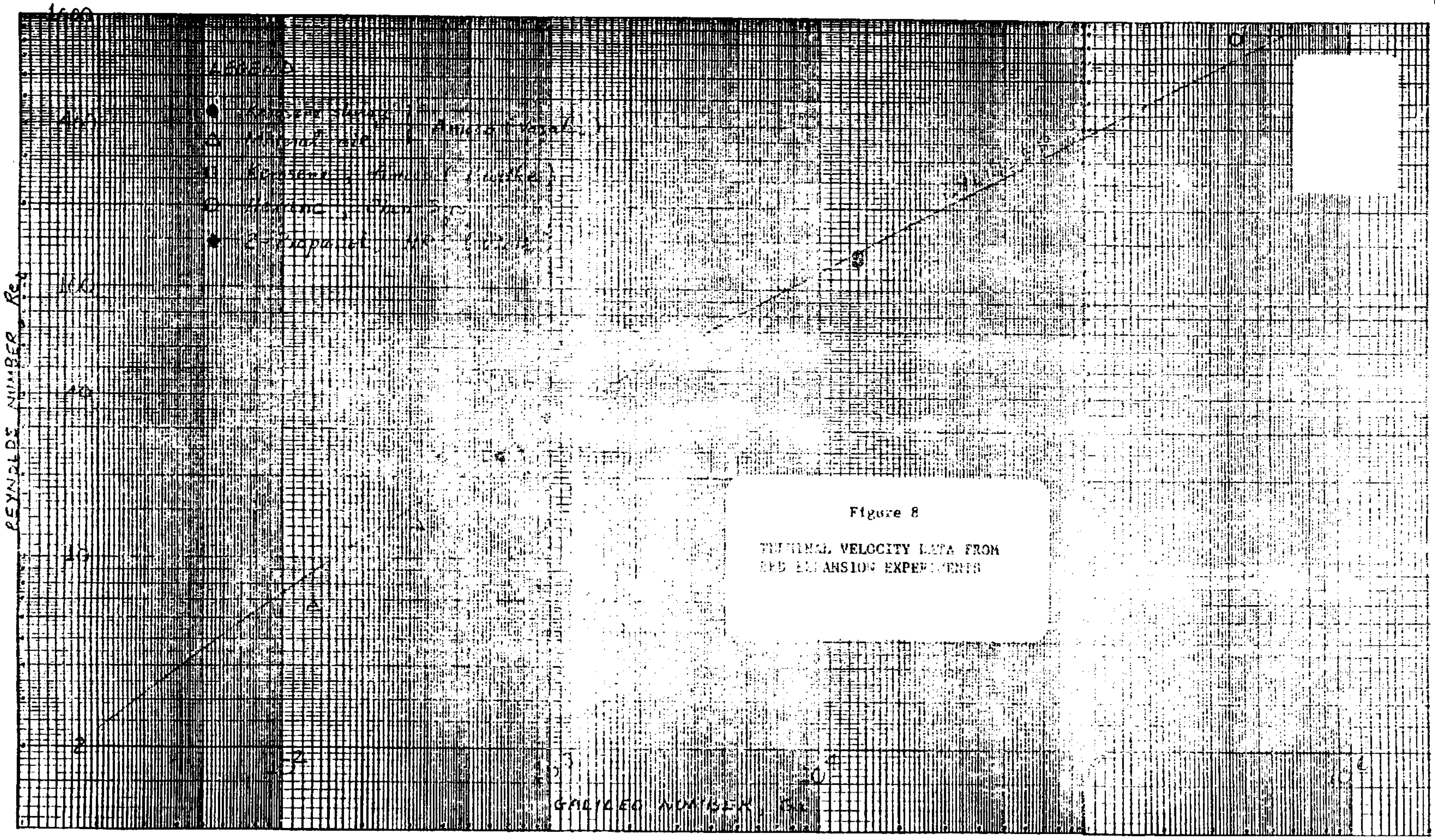


Figure 8

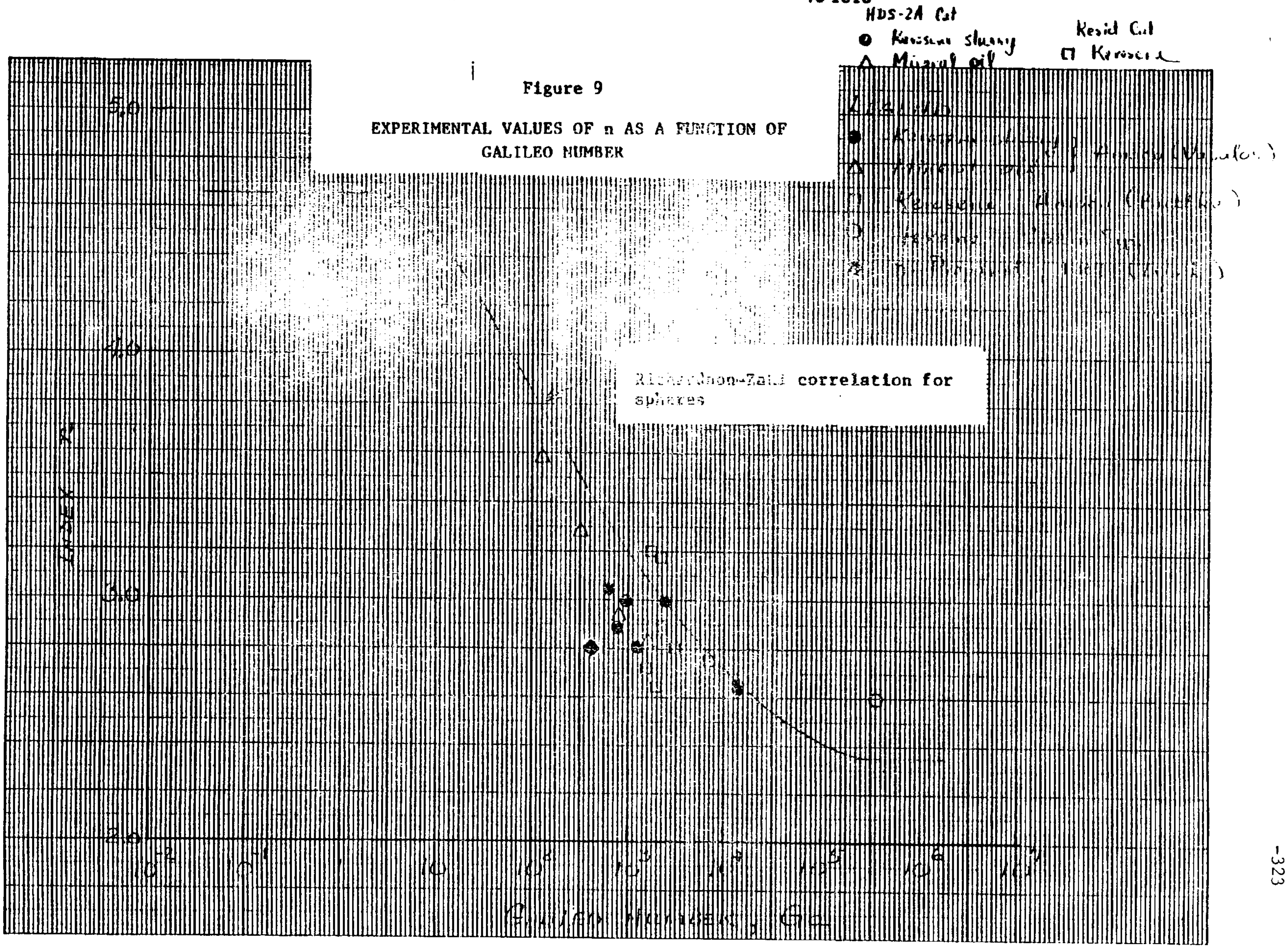
TERMINAL VELOCITY DATA FROM BED EXPANSION EXPERIMENTS

K•E 10 X 10 TO THE CENTIMETER 18 X 25 CM.
KEUFFEL & ESSER CO. MADE IN U.S.A.

46 1513

Figure 9

EXPERIMENTAL VALUES OF n AS A FUNCTION OF
GALILEO NUMBER



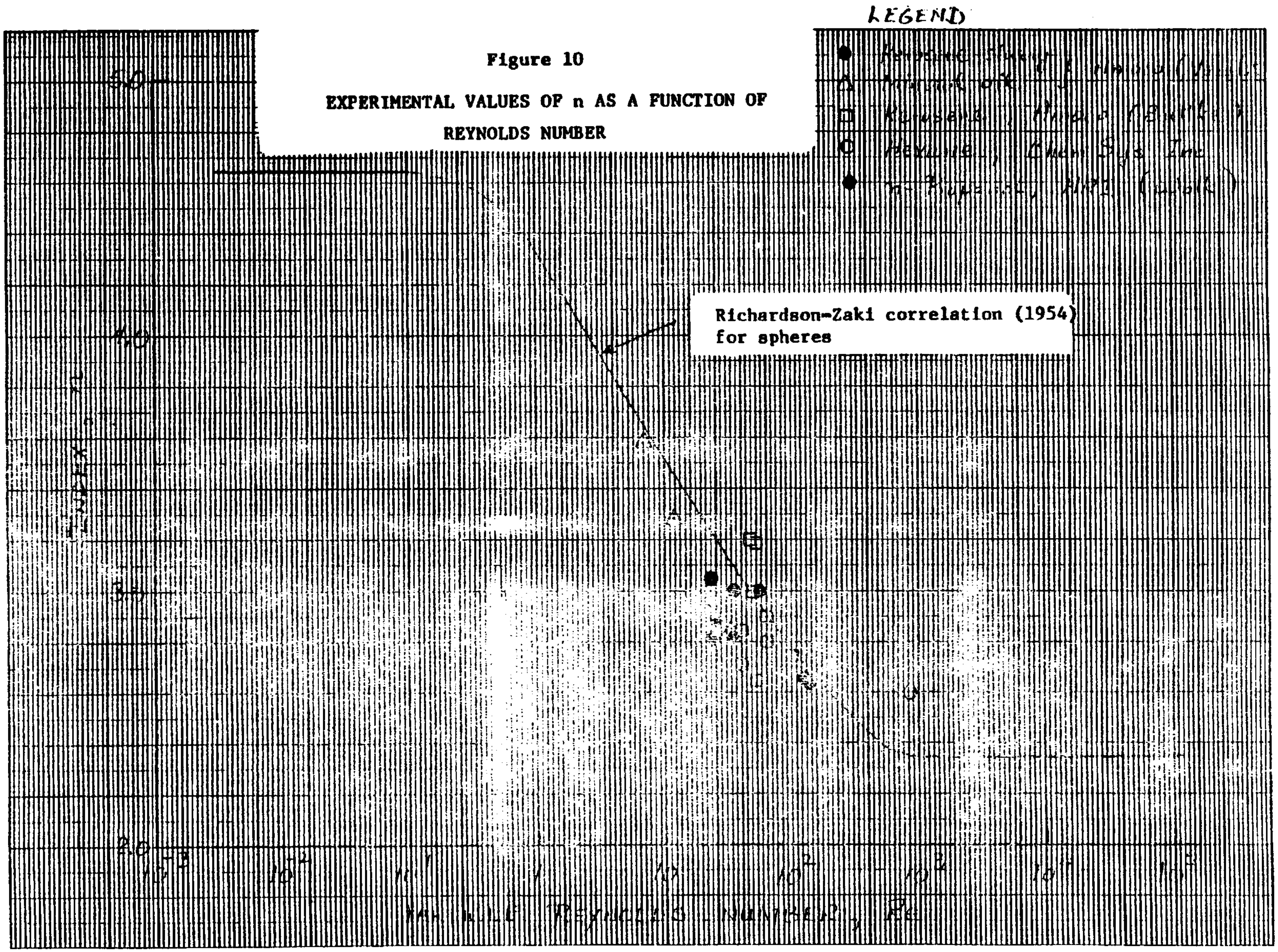


Figure 10

EXPERIMENTAL VALUES OF n AS A FUNCTION OF REYNOLDS NUMBER

APPENDIX

RICHARDSON-ZAKI PLOTS OF BED EXPANSION DATA

The Richardson-Zaki plots for the bed expansion data are presented in this appendix.

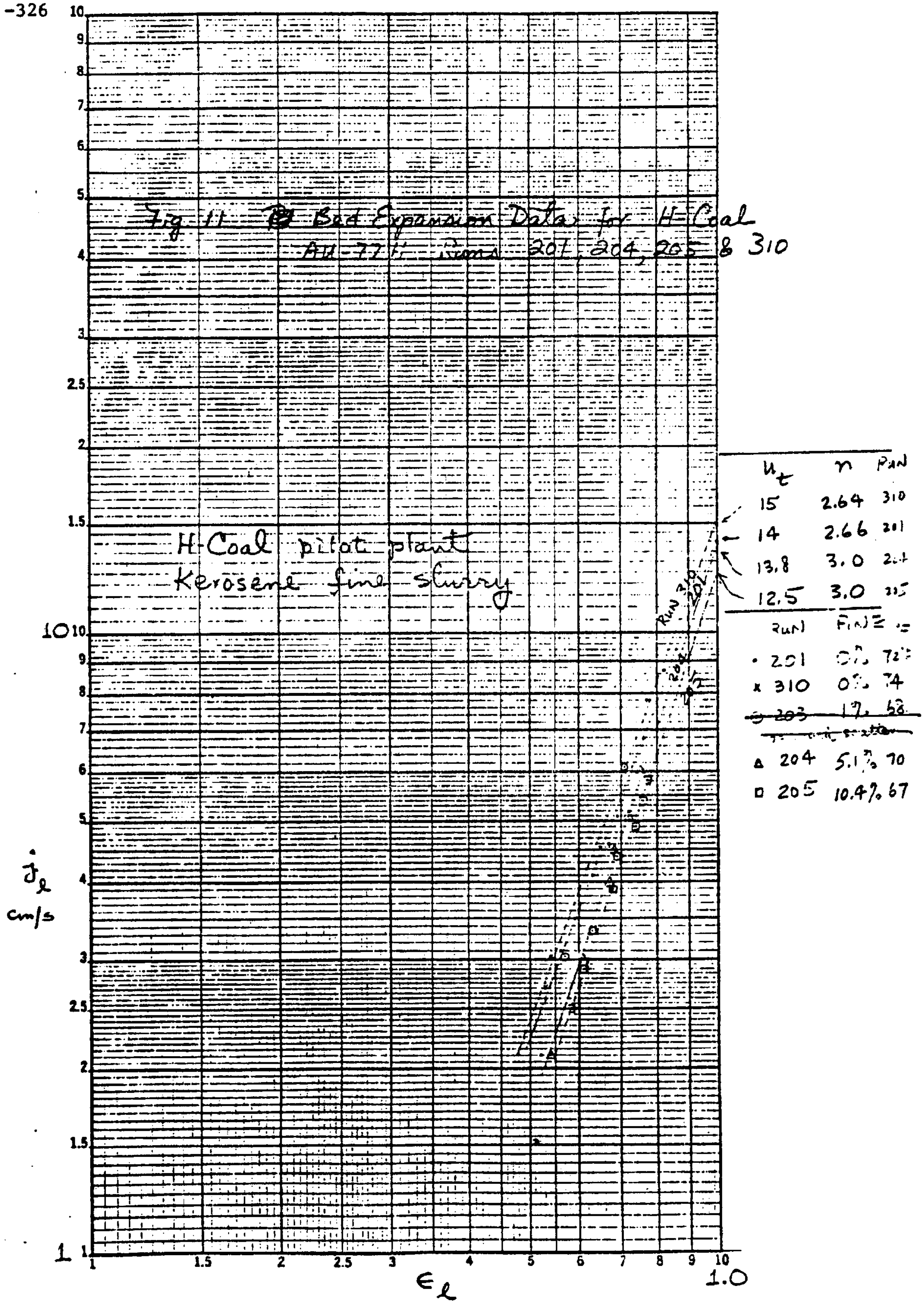


Fig. 11 Bed Expansion Data for H-Coal
All 77°F Runs 201, 204, 205 & 310
H-Coal pilot plant
Kerosene fine slurry
u_t n RUN
15 2.64 310
14 2.66 201
13.8 3.0 204
12.5 3.0 205
RUN FINES
• 201 0% 72°F
x 310 0% 74
△ 204 5.1% 70
□ 205 10.4% 67
RUN 310
201
j_ℓ
cm/s
ϵ_ℓ
1.0

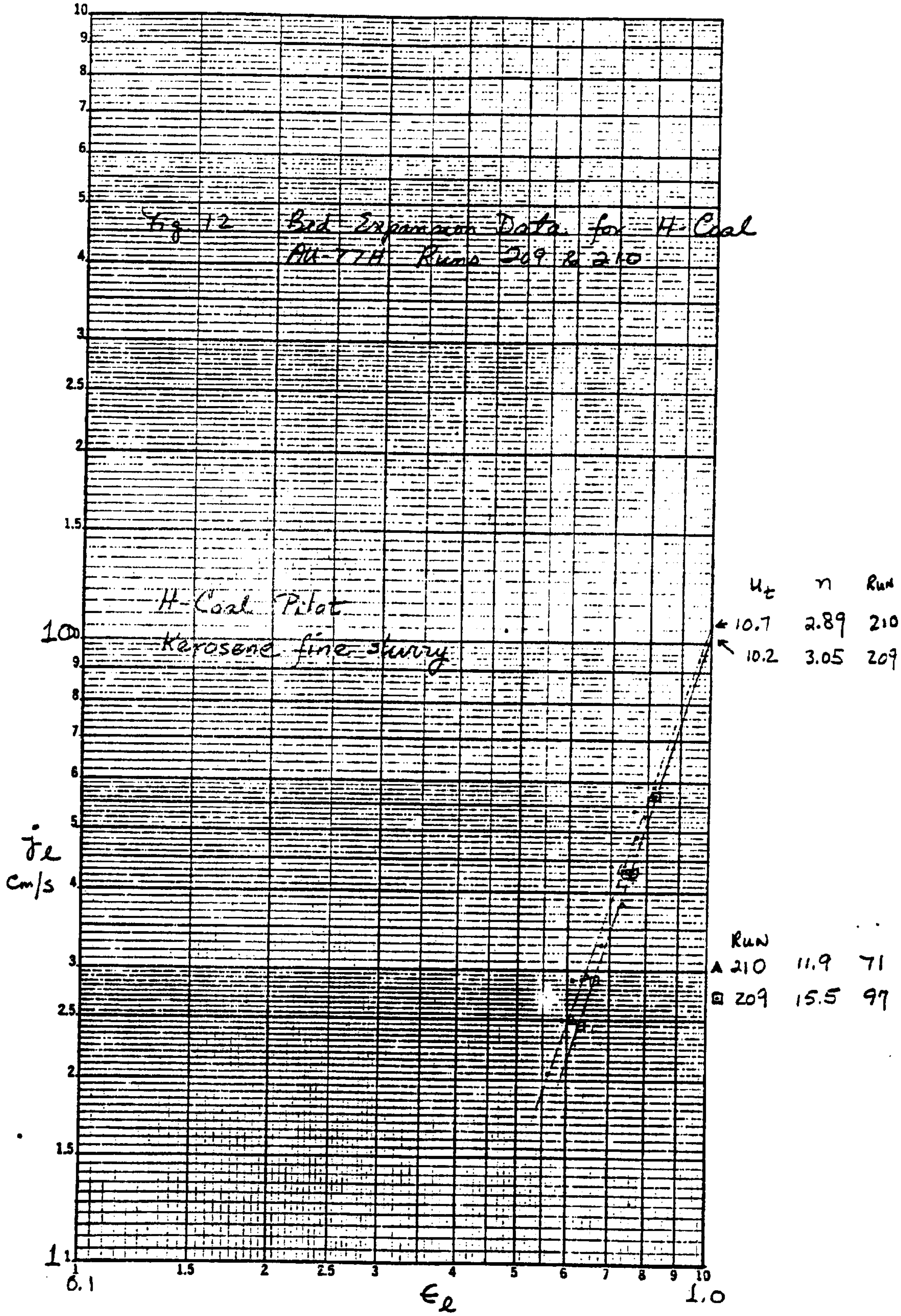


Fig 12 Bed Expansion Data for H-Coal
AU-77H Runs 209 & 210
H-Coal Pilot
Kerosene fine slurry
u_t n Run
10.7 2.89 210
10.2 3.05 209
Run
▲ 210 11.9 71
⊡ 209 15.5 97
j_l cm/s
ε_l
0.1
1.0
1
10
100

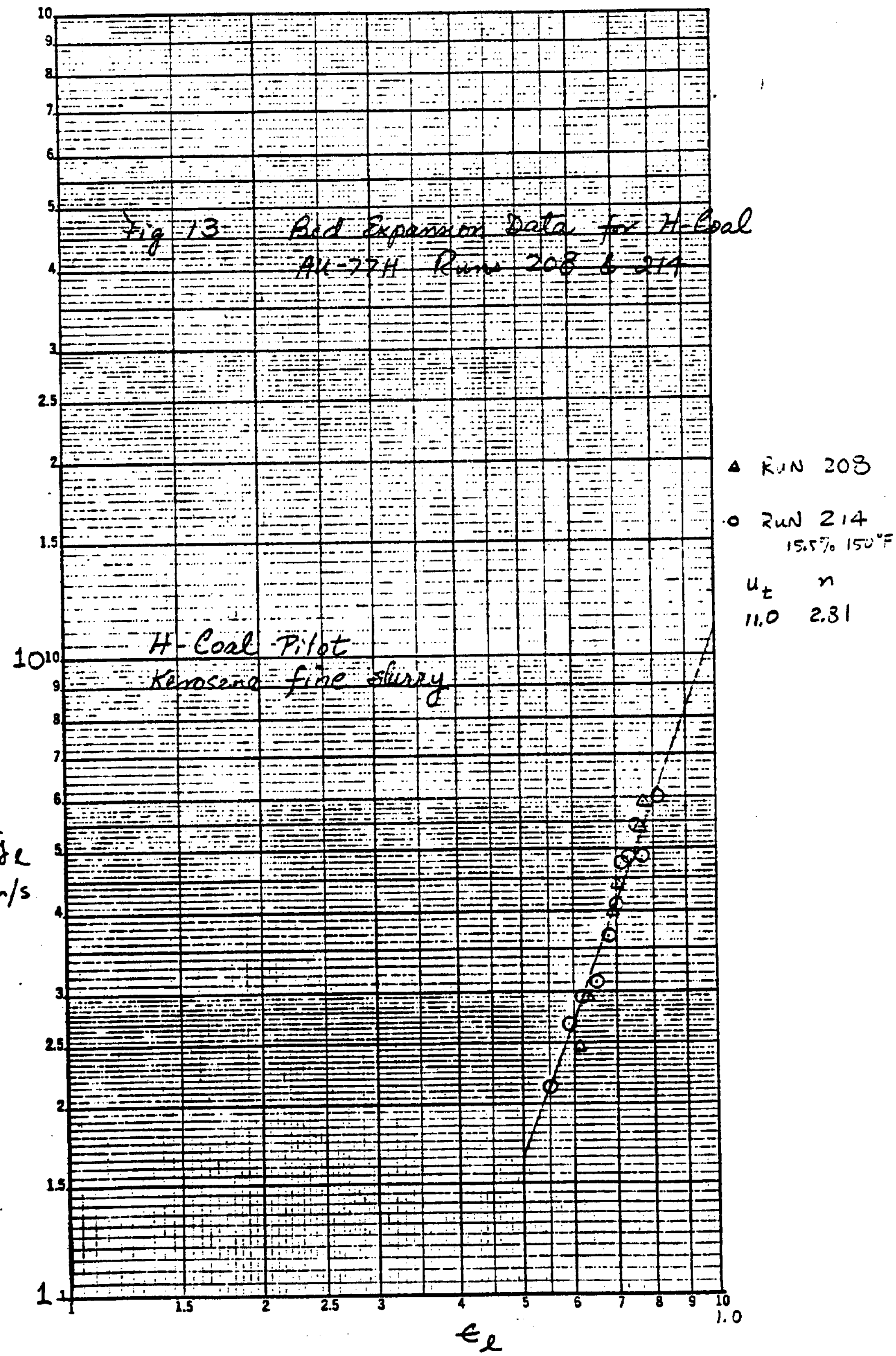


Fig 13 Bed Expansion Data for H-Coal
AU-77H Runs 208 & 214
H-Coal Pilot
Kerosene fine slurry
▲ RUN 208
○ RUN 214
15.5% 150°F
u_t n
11.0 2.31
j_ℓ
cm/s
ϵ_ℓ

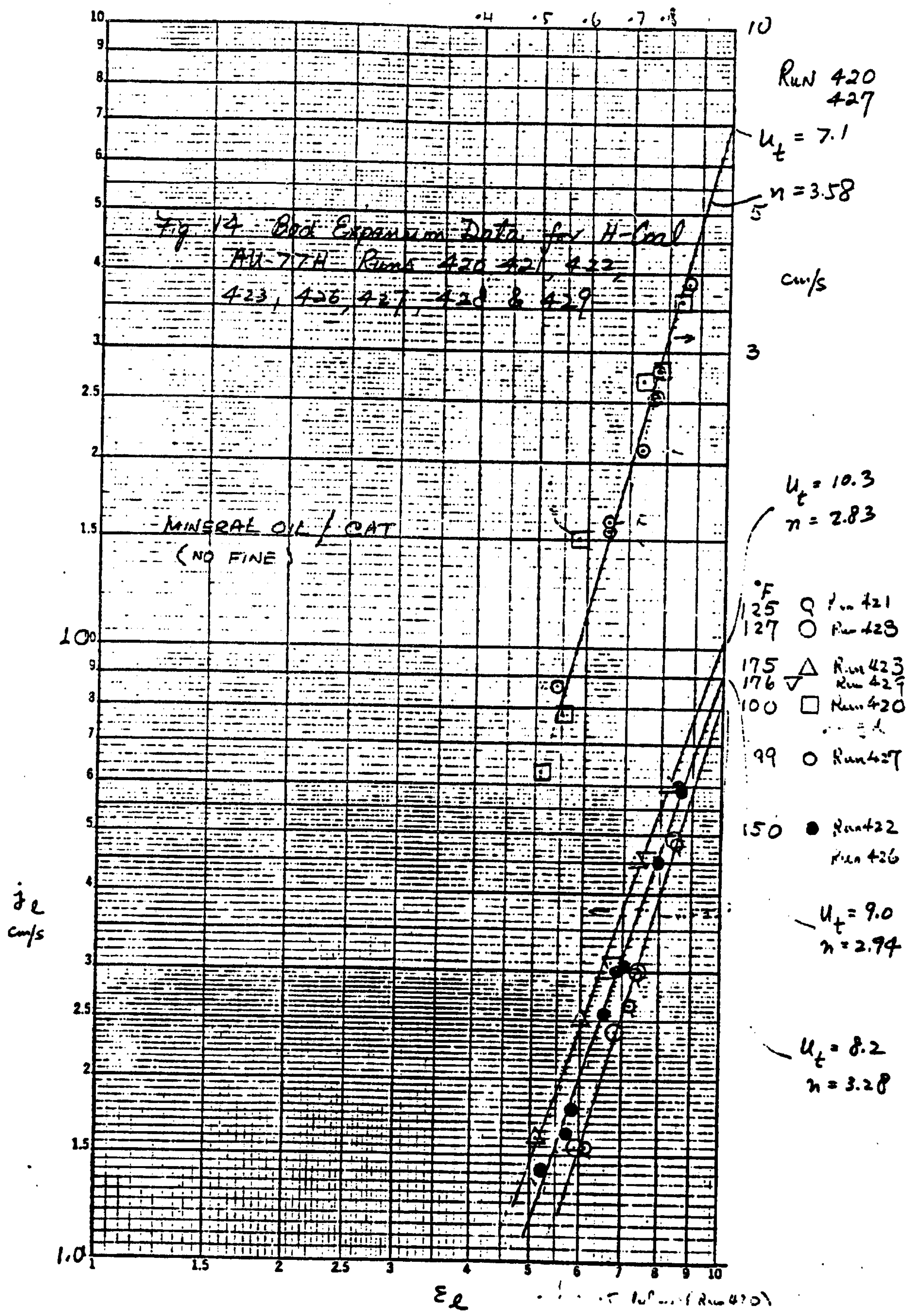
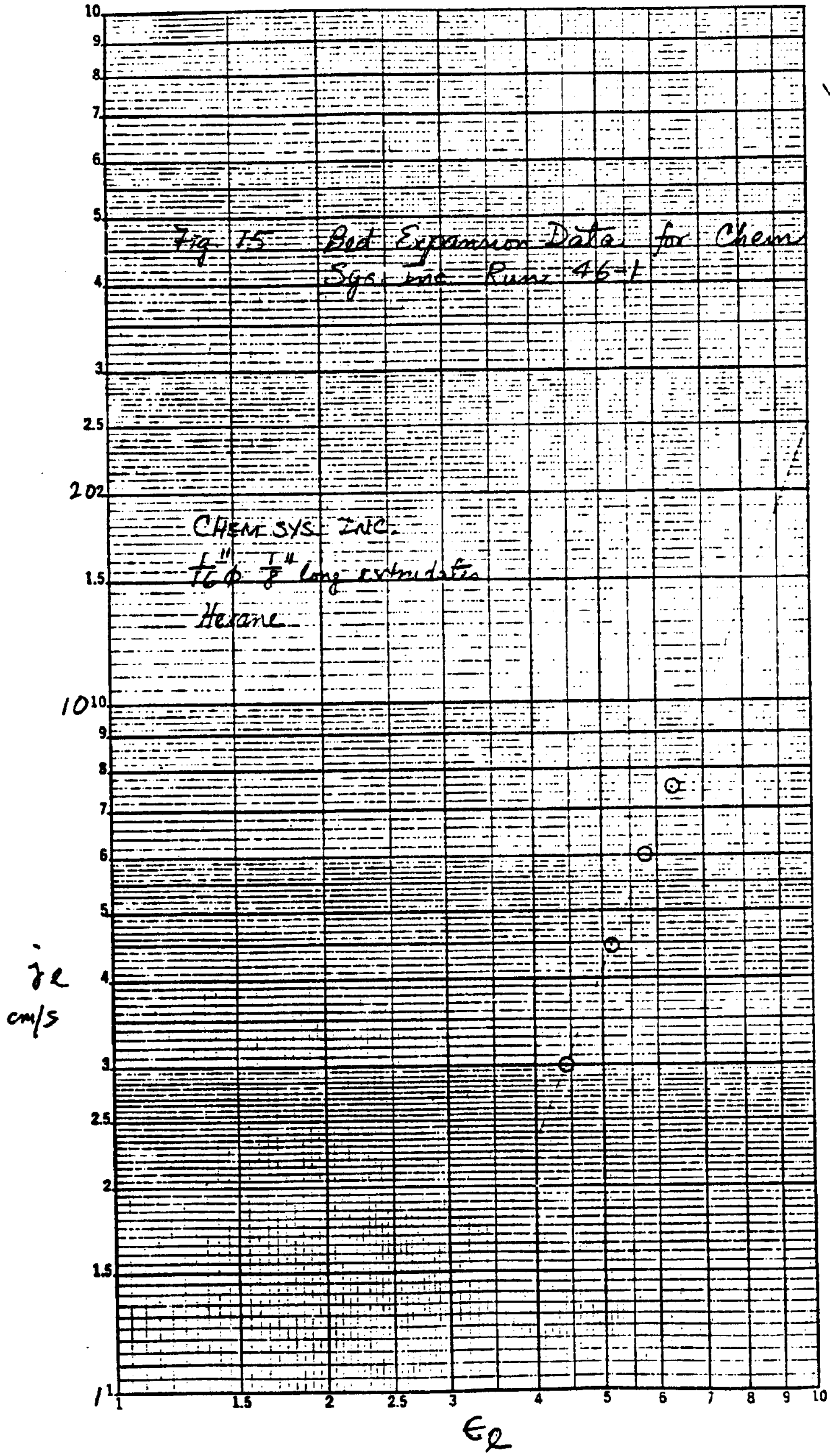


Fig 14 Bed Expansion Data for H-Coal
AU-77H Runs 420 421, 422
423, 426, 427, 428 & 429
MINERAL OIL / CAT
(NO FINE)
RUN 420
427
u_t = 7.1
n = 3.58
cm/s
u_t = 10.3
n = 2.83
°F
125 Run 421
127 Run 428
175 Run 423
176 Run 429
100 Run 420
99 Run 427
150 Run 422
Run 426
u_t = 9.0
n = 2.94
u_t = 8.2
n = 3.28
j_ℓ
cm/s
ε_ℓ

VCL 9/13/81

u_t 25 cm/s
n 2.6



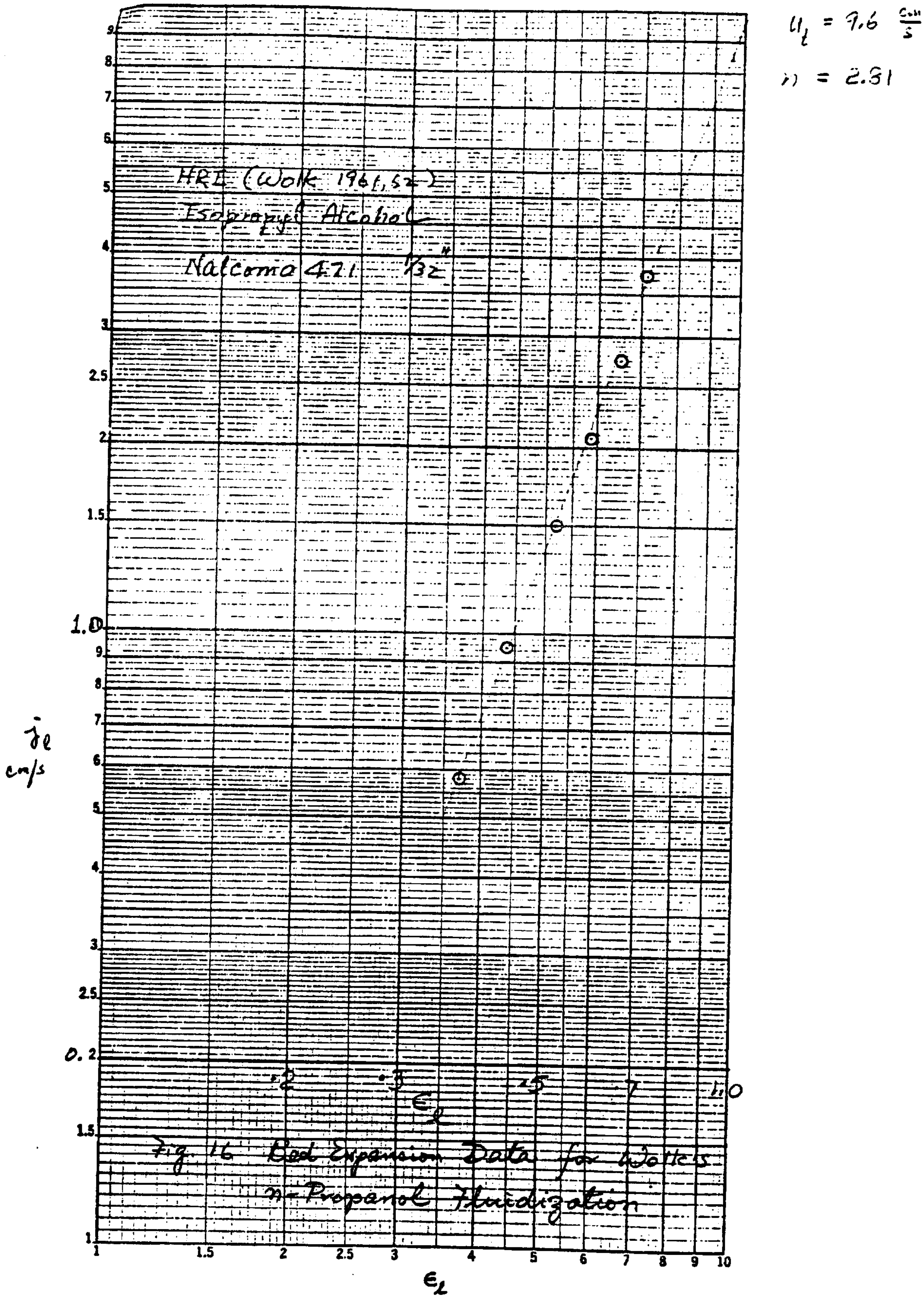


Fig. 16 Bed Expansion Data for Wolk's n-Propanol Fluidization