FLUIDIZATION PROPERTIES OF COAL CHAR

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INTRODUCTION

Coal reserves are vast and therefore will play an ever-increasing role in supplying our growing energy demands. However, because of predicted shortages of liquid and gaseous fuels and because of environmental considerations, future utilization will include the development of processes to desulfurize coal as well as to convert it to liquid and gaseous fuels that would also be desulfurized.

Thus, processes such as pyrolysis, desulfurization, gasification, hydrogasification, carbonization, as well as fluid-bed combustion, are being developed to allow coal to broaden its role in satisfying future energy and chemical demands. These processes have at least two features in common: First, they will likely utilize char, derived from coal, as a feed or a constituent in some reactor zone. Second, as a corollary of the particulate nature of char, the processes can use fluidized-bed reactors to take advantage of the improved heat and mass transfers, easier flow operations, good mixing, and ease of control. The design of reactor systems to carry out the above processes therefore requires knowledge of the fluidization properties of coal chars, as well as knowledge of reaction thermodynamics and kinetics.

This study was made to provide the necessary fluidization data for the design of fluid-bed hydrogasifiers of both laboratory and commercial scale. An excellent summarization of fluidization data for various materials appears in the literature.[1] Additional data on chars are also available.[2,3] However, the char we will be fluidizing is so different that use of the published data is precluded. The correlations presented here should allow extension of data to the design of any other fluid-bed reactor system using coal chars that have the same physical characteristics. Apparently, no universal correlation has been developed to account perfectly for all the physical parameters involved in fluidization; thus, no single correlation now exists that suits all particulate materials regardless of constitution or especially particle shape. Separate correlations still appear to be needed for widely varied materials, as will be demonstrated here. The particular char used in these experiments was produced by rapid hydrogasification of raw high-volatile bituminous coal in a "free-fall" reactor. In our overall process concept to produce pipeline gas from coal, this lightly converted char (25 to 30 percent carbon conversion) would fall into a fluid-bed reactor for additional carbon conversion and for the removal of sufficient hydrogasification heat to maintain the reaction temperature at 1,600° to 1,700° F. The free-fall reactor system has been analyzed[4] and an integrated reactor system is now being designed to add a fluid-bed reactor below this dilute phase to allow both an experimental and an engineering evaluation of the overall process thus integrated.

EXPERIMENTAL

Material. The char used in this study was produced by concurrent injection of raw bituminous coal and hot hydrogen:methane (1:1) mixture into the top of a 5 ft x 3 in. id reactor at 1,000 psig and 1,650° F. Very rapid devolatilization, gasification of the volatiles, and partial hydrogasification of char all occur. The resultant char falls freely in the slowly down-flowing gas. Carbon conversion in the above reactor is from 25 to 30 percent.

Apparatus. The fluidization studies were conducted at atmospheric pressure with the majority of the tests in a plexiglass column of 3.69-inch id. The fluidizing gas was introduced at the bottom of the column through a porous sintered stainless steel distributor plate, 1/4 inch thick with an average pore size of 5 microns. Thinner, more porous distributors were avoided when found to induce more channeling in the fluid bed, especially at the reactor bottom. The bed depth was varied from almost 3 feet to a few inches with no appreciable change in fluidization properties. Two experiments were made in a two-inch id tube with static heights of approximately five feet to observe the fluidization behavior of this char at conditions anticipated for a bench-scale fluidized-bed char hydrogasifier. Gas rates and pressure drops were measured with the usual rotameters and manometers.

RESULTS

Char Properties. A char property important to fluidization is the effective particle density. Because of the sudden heating in the dilute-phase hydrogasifier as described above, this char has a very large fraction of internal void space so that the density is much less than that of the feed coal or chars from slow carbonization. A photomicrograph of a typical char particle is shown in figure 1 to illustrate the internal structure of this char.

Because of the vesicularity of this char, the effective density of the char particles is not known directly, but can be calculated from bulk density and gas-flow pressure drop measurements made on packed beds according to the procedure recommended by Leva[5] using Ergun's correlations.[6]

As shown in figure 2, the particle densities measured in the above manner fall into two categories, depending on the particle diameter. For fine particles where $\overline{d}_p$ <.003 in., the effective particle density is about 26 lb/cu ft while for the coarser particles where $\overline{d}_p$>.01 in., the particle density is about 16 lb/cu ft. For .003 < d_p <.01 there seems to be a transitional region, although only one date point is included in this particle size range. The most likely explanation of this behavior is that below a certain particle diameter, the particles cannot be vesicular and therefore must have a higher effective solid density than larger particles which can contain many vesicules. This explanation is consistent with the microphotograph shown in figure 1 because the approximate size range of the vesicules is about equal to the particle diameter at which the density transition occurs. Therefore, because the vesicules seem to have a limiting minimum size, particles smaller than this minimum size are nonvesicular. Physical interpretation of this effect is possible. Volatile matter evolved inside small particles of rapidly heated coal can escape without forming a vesicule because of the shorter diffusional path, or because any "skin" of coal in its plastic phase is too thin and bursts before a sizable vesicule can form. The larger particles can have thicker, stronger plastic layers to contain the evolved volatile matter long enough to make vesicules, and the diffusional escape of gas is, of course, more restrained.

The bulk density dependence on mean particle size is shown in figure 3. The mean particle diameter is here defined, as recommended by Leva,[4] by $\frac{1}{\overline{d}_p} = \Sigma \frac{Xi}{d_{pi}}$. The discontinuity of bulk density versus size occurs at the same particle diameter as was observed for the discontinuity of particle density versus size. However, the bulk density curve for the vesicular particles is slightly above the curve for the nonvesicular particles in spite of the higher solid density of the nonvesicular particles. The most likely explanation for this experimental observation is that the wider variety of particle shapes one finds in the larger particle size mixtures permits closer packing which more than compensates for the decreased solid density.

The smaller sized particles, being more spherical, apparently pack with a higher void space, causing a decrease in the bulk density.

Minimum Fluidization Velocity. The minimum fluidization velocity was found to depend on the properties of the solid and fluid in a way that permits use of the dimensionless Galileo number, N_{Ga} for correlation. A correlation that empirically fits the data was obtained by plotting log (N_{Re}) against log (N_{Ga}), as suggested by Wen and Yu.[1] The resulting correlation is shown in figure 4. An excellent straight line results for both narrow and wide particle size distributions when the mean particle diameter as defined above is used.

Some other salient features of the correlation presented in figure 4 are: (1) Though self-consistent, our correlation using mean particle sizes does not agree with that of Wen and Yu.[1] (2) For narrow size distributions, as expected, it does not make much difference whether mean or maximum particle size is used; a self-consistent correlation occurs either way. (3) For the few points available for broad size distributions, attempted correlation is poor when maximum particle size, as recommended by Wen and Yu, is used instead of mean particle size.

In view of the above three features, it is recommended that average particle diameters be used with our correlation for predictive purposes with similar chars.

In general, the chars used in these studies require much larger velocities to fluidize than predicted by reference 1. The difference between the correlations is too great to attribute to inaccuracies in measuring ρ_s which is the least accurate term in the Galileo number. Examination of the char particles reveals the most likely reason for the deviation between correlations is the configuration or shape of the respective particles. A picture of some typical char particles in figure 5 shows that the "particles" consist of a sizable fraction of agglomerates of small particles and that these agglomerates can readily interlock. Thus, collisions between particles in the fluid bed will frequently result in the particles catching together which results, momentarily, in an increase in the effective particle diameter in the bed and the requirement of additional energy to move the combination or to unhook the particles. Because this additional energy can only be supplied through increased gas velocity, these particles fluidize at higher velocities than particles having identical properties except for less ability to interlock upon collision. In deciding whether to use the correlation based on these results or that based on more "regular" particles, it would probably be sufficient to examine the particles to see whether they contain shapes that can cause them to hook together and thereby require the use of higher fluidization velocities.

A summary of the fluidization data in given in table 1 to illustrate the range of particle sizes and bed heights used to derive the correlation in figure 4. The minimum fluidization velocity was obtained in the classical manner by plotting the experimentally measured bed pressure drop against the gas velocity, and defining the velocity at which ΔP just reaches $\Delta P = \frac{W_s}{A_T}$ as the minimum fluidization velocity.

A typical ΔP vs u curve is shown in figure 6a for a relatively narrow particle size range.

In general, fluidization was visibly smoother for narrow size fractions of particles. However, wide fractions of only coarser particles also behaved well. The most irregular fluidization was experienced with wide size fractions having appreciable concentrations of fine particles, leading to behavior illustrated in figure 6b,

TABLE 1. - Summary of fluidization data for coal char fluidized with CO_2; tube id = 3.69 (with exceptions noted below)

Particle size range, U. S. Sieve Series	Mean particle diameter, in.	Minimum fluidization velocity, ft/sec	Reynold's No. at minimum fluidization	Galileo number	Bed height at minimum fluidization, in.	Wt of char in bed, lb
6 x 8	0.113	1.42	151	3.6×10^5	9.0	0.248
8 x 12	0.08	1.06	80	1.2×10^5	11.0	0.339
12 x 14	0.061	0.74	43	6.24×10^4	8.2	0.280
14 x 20	0.044	0.57	23.8	2.5×10^4	10.8	0.410
20 x 40	0.025	0.37	8.8	5.15×10^3	10.6	0.480
40 x 60	0.013	0.14	1.8	810	3.7	0.209
60 x 80	0.0084	0.11	0.87	254	2.6	0.183
80 x 120	0.0059	0.085	0.48	101	2.0	0.163
120 x 200	0.0039	0.059	0.21	28.6	11.6	0.789
200 x 325	0.0022	0.040	0.080	6.02	13.0	1.51
1/ 6 x 40	0.065	1.23	75	1.34×10^5	24.0	0.546
1/ 6 x 40	0.071	1.27	85	1.57×10^5	30.0	0.825
1,2/ 100% -14, 34% -325	0.0052	0.05	0.25	61	47.0	1.0
1,2,3/ 100% -14, 34% -325	0.0052	0.03	0.076	26	47.0	1.0

1/ Data given by solid points on figure 4.
2/ Tube id = 2 inches.
3/ Fluidized with N_2.

where precise definition of U_{mf} was difficult. Appreciable channelling and segregation was noted with the particle size range illustrated in figure 6b, but this could be overcome by going to velocities considerably higher than U_{mf} where performance stabilized even with the relatively high L/D ratio used in this test.

Design Considerations. To design a fluid-bed chemical reactor, it is generally not sufficient to know only U_{mf} because one must operate at a velocity sufficient to give vigorous mixing and prevent the segregation of particles. With the narrow particle size ranges, it was found that a gas velocity twice that of U_{mf} was sufficient to provide good mixing and to prevent segregation. However, as mentioned above, for the wide particle size distribution (100 percent -14 to 34 percent -325) in table 1, it was necessary to operate at $U/U_{mf}>7$ before complete mixing was achieved.

To calculate the mean solids residence time in a fluid-bed reactor requires knowing the amount of bed expansion as a function of operating conditions. The data below indicate that the expansion ratio depends on the mean particle size as well as U/U_{mf}. A summary of experimental results is shown in figure 7a and 7b where L/L_{mf} is plotted against U/U_{mf} for various particle sizes. As the mean particle size decreases, so does the bed expansion ratio for a fixed U/U_{mf} until d_p reaches about 0.025 in., below which further reductions in particle size do not change the expansion behavior. For wide particle size ranges, the previously defined mean diameter determines the bed expansion as is indicated in figure 7b where expansion ratios for fluid beds having char particles from 14 to325 mesh are plotted. Also in figure 7b, data are shown for narrower particle size distribution tests made with particle diameters below .025 inch.

CONCLUSIONS

The minimum fluidization velocity of coal chars, physically similar to those studied here, can be predicted for various particle sizes and gas conditions by computing the Galileo number and fitting it to the correlation developed here. To get the Galileo number requires knowledge of mean particle size, particle density, and gas density and viscosity at conditions for the desired system. The correlation applies to both narrow and wide particle size distributions and over a range of "Reynolds numbers at minimum fluidization" from less than 0.1 to over 100. The minimum fluidization velocity predicted by our experimental correlation is considerably higher than one would calculate from existing theory and from other correlations because of small agglomerates in the char which causeentanglement and thus require an additional energy input to either separate them or fluidize the more massive combination. Correlations are also presented for fluid-bed expansion that indicate that the mean particle size is the main factor in determining how bed expansion will vary with gas velocity.

NOTATION

U = superficial gas velocity, ft/sec

U_{mf} = superficial gas velocity, at minimum fluidization, ft/sec

ρ_F = density of fluid phase, lb/cu ft

ρ_s = effective density of solid phase, lb/cu ft

ρ_b = bulk density of bed, lb/cu ft

d_{pi} = particle diameter, ft

Xi = weight fraction of particles having diameter d_{pi}

d_p = mean particle diameter, ft

μ = gas viscosity, lb/ft-sec

W_s = weight of solid in bed, lb

A_T = cross sectional area of tube, sq ft

ΔP = pressure drop across bed, lb/sq ft

L = bed height, ft

Lmf = bed height at minimum fluidization, ft

D_T = tube diameter, ft

g = acceleration of gravity, ft/sec^2

N_{Ga} = Galileo number = $d_p \rho_f (\rho_s - \rho_f)\, g \mu^{-2}$

(N_{Re}) = Reynold's number at minimum fluidization

$= d_p\, U_{mf}\, \rho_f\, \mu^{-1}$

REFERENCES

1. Wen, C. Y. and Y. H. Yu. Chem. Eng. Prog. Symposium Series, v. 62, No. 62, 1966, pp. 100-111.

2. Jones, J. F., R. T. Eddinger, and L. Seglin. Proceedings of International Symposium on Fluidization, Eindhoven Netherlands, N.U.P. Amsterdam, June 6-9, 1967, published by Netherlands University, Amsterdam.

3. Curran, G. P., and Everett Gorin. Studies on Mechanics of Fluo-Solids Systems. Period July 1964 to June 1968, R & D Report No. 16, Interim Report No. 3, Book 1, OCR Contract 14-01-0001-415, Consolidation Coal Co., Research Division, Library, Pa.

4. Feldmann, H. F., W. H. Simons, J. A. Mima, and R. W. Hiteshue. ACS Div. of Fuel Chem. Preprints, v. 14, No. 4, Part II, pp. 1-13, Sept. 1970.

5. Leva, Max. Fluidization, McGraw Hill Book Co., Inc., N. Y. 1959, pp. 60, 61.

6. Ergun, Sabri. Anal. Chem., v. 24(2):388 (1952).

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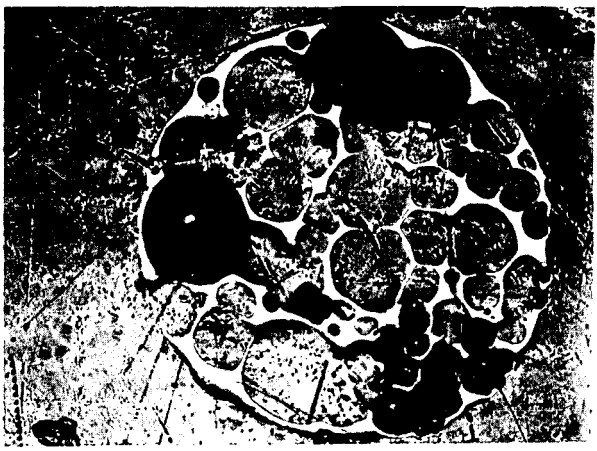

FIGURE 1.- Cross Section of Typical Char Particle at 220 Magnification.

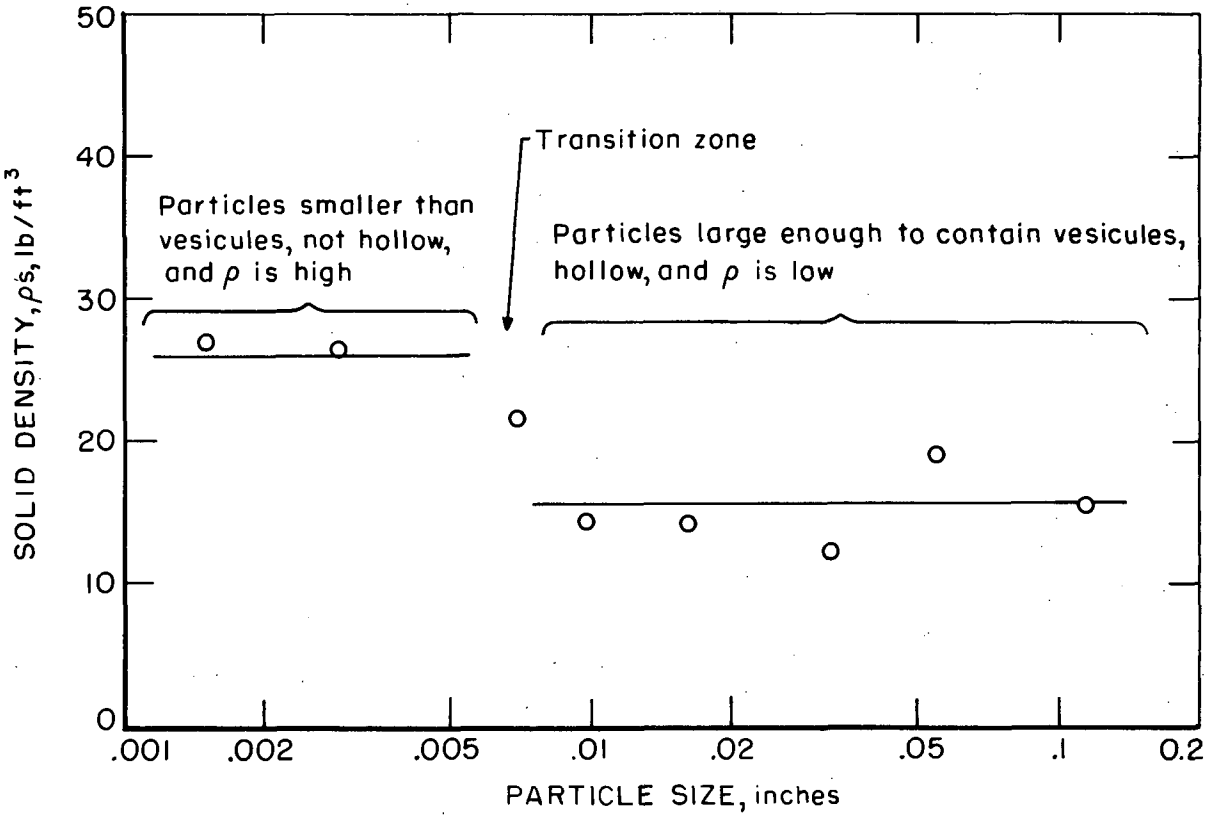

FIGURE 2.- Particle Density vs Particle Size of Char.

L-12230

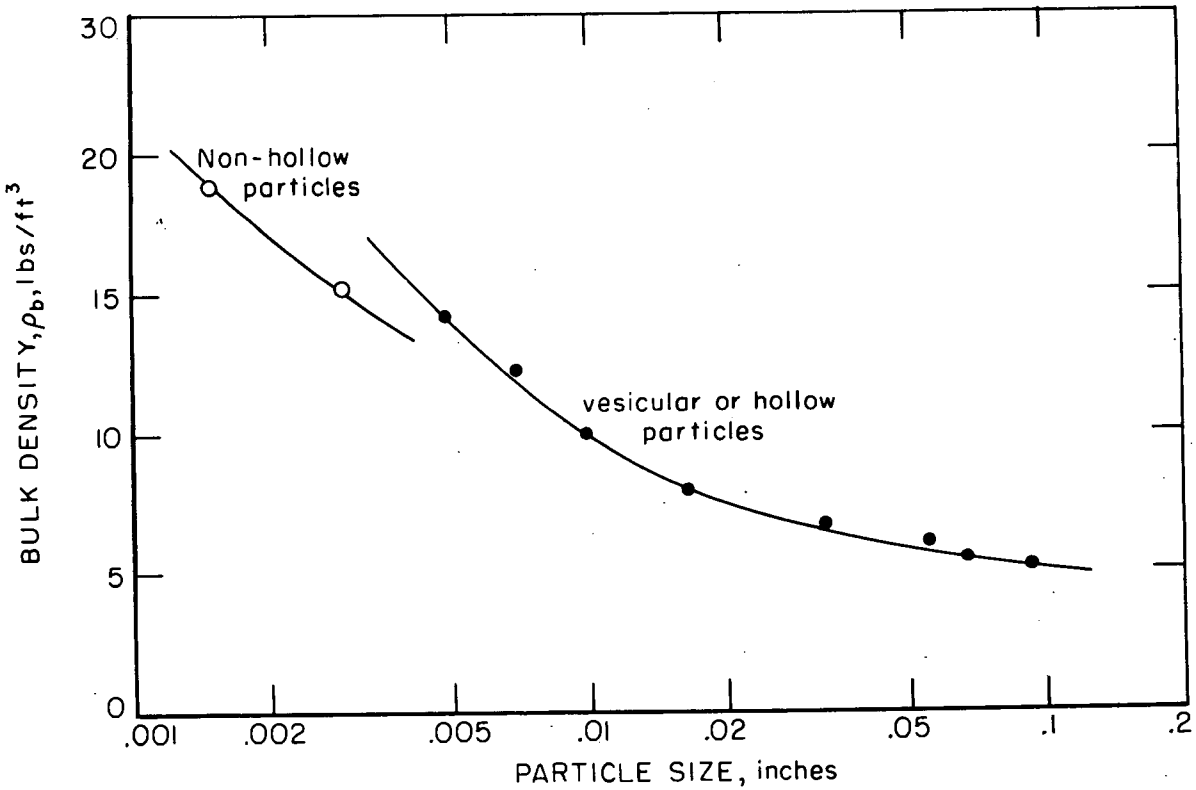

FIGURE 3.- Bulk Density vs Particle Size of Char.

L-12231

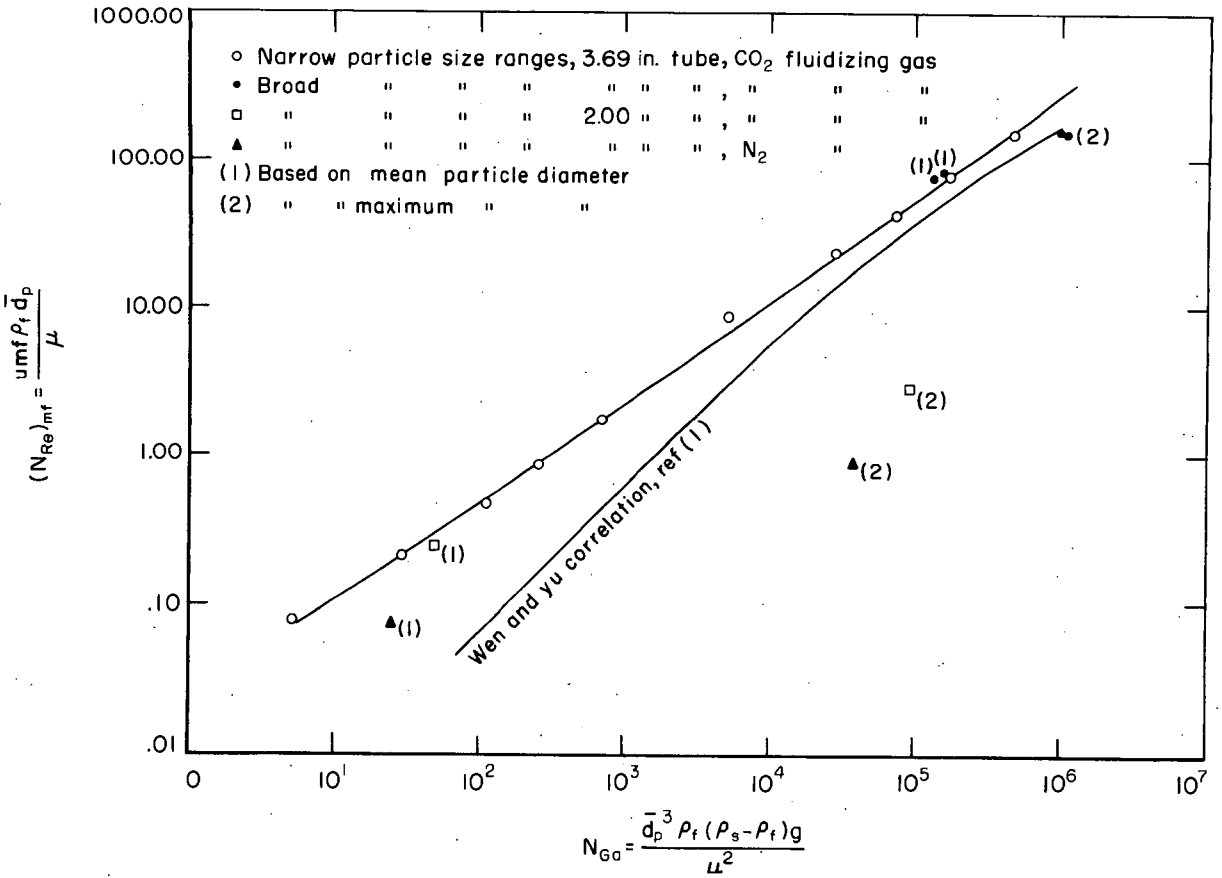

FIGURE 4.- Fluidization Correlation.

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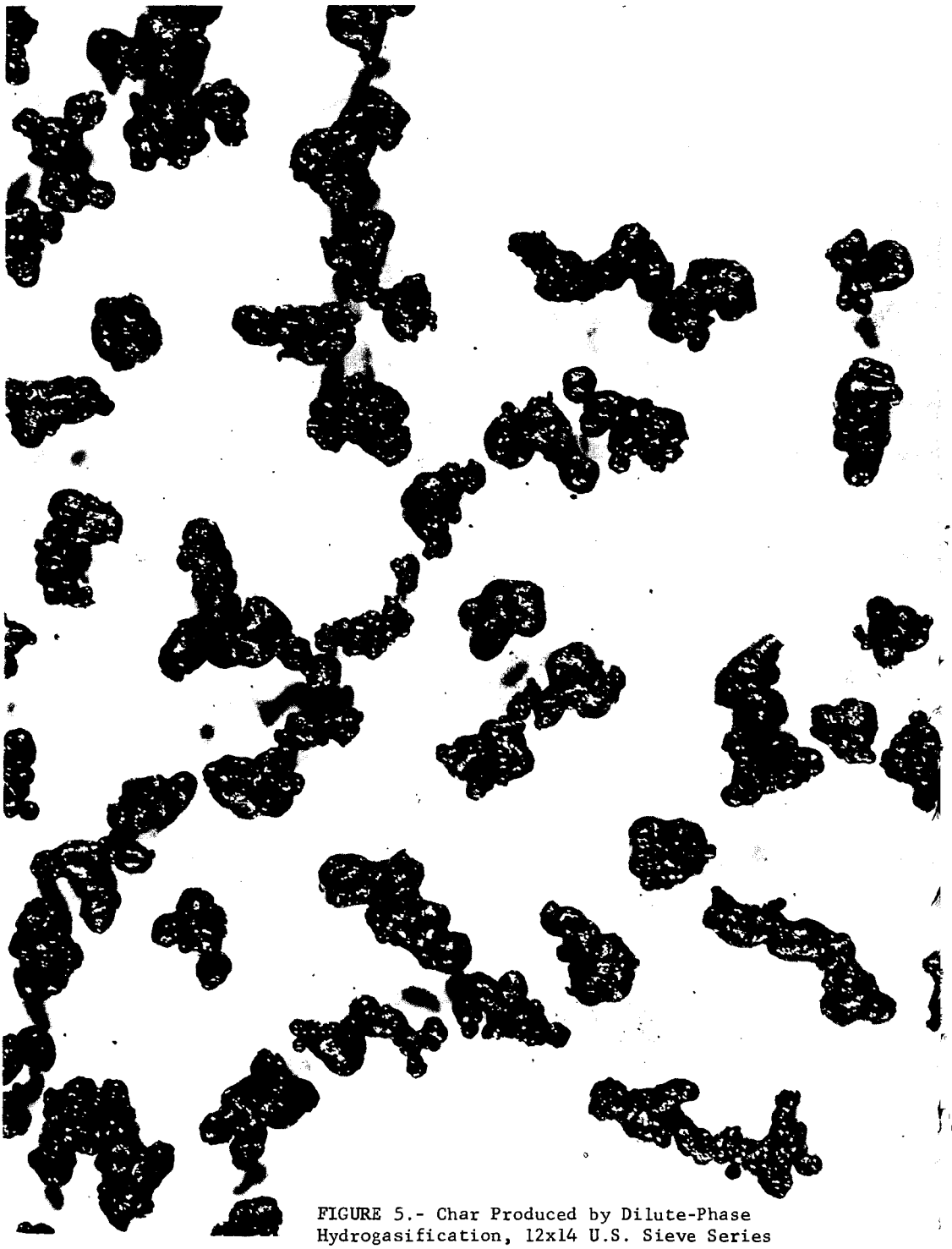

FIGURE 5.- Char Produced by Dilute-Phase Hydrogasification, 12x14 U.S. Sieve Series

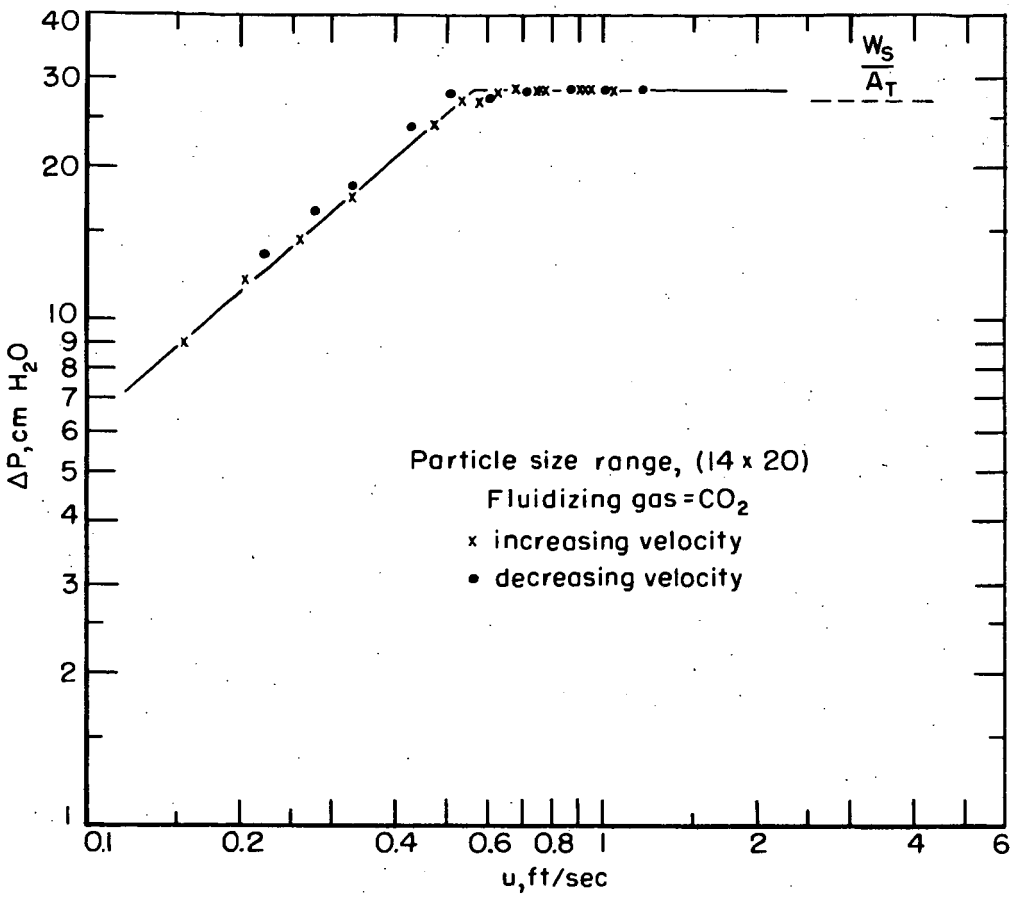

FIGURE 6a.- Typical Pressure Drop-Velocity Curve, Narrow Particle Size Range.

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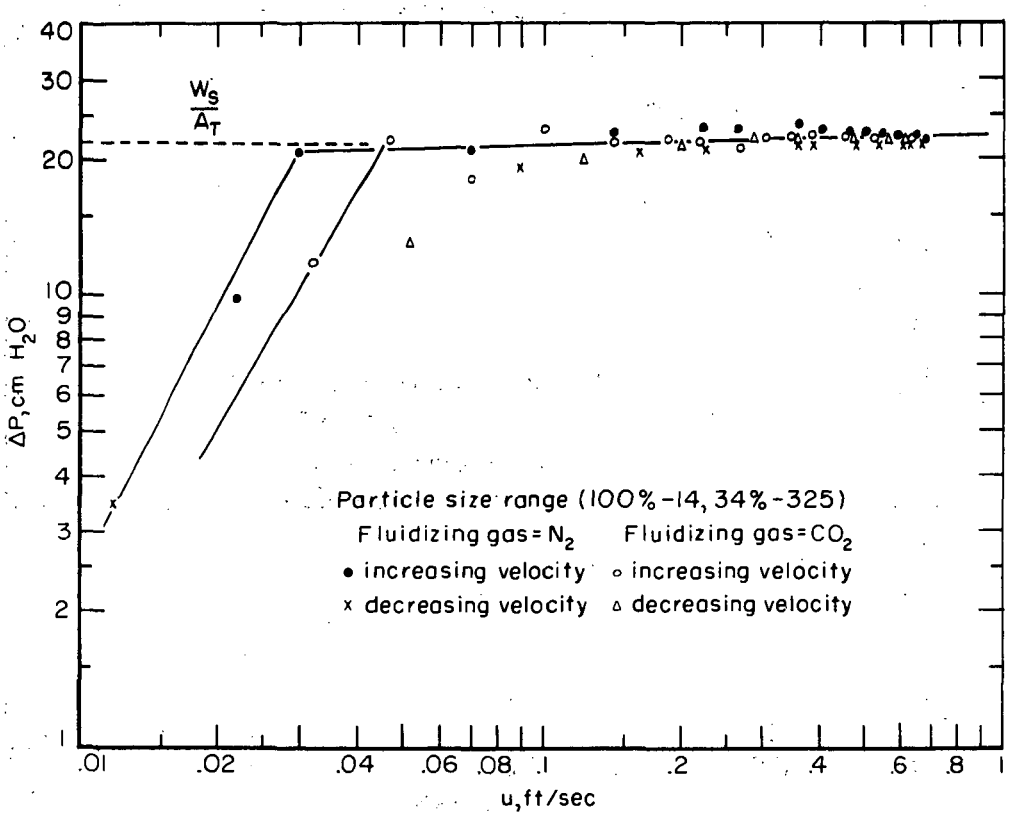

FIGURE 6b.- Typical Pressure Drop-Velocity Curve, Broad Particle Size Range.

L-12234

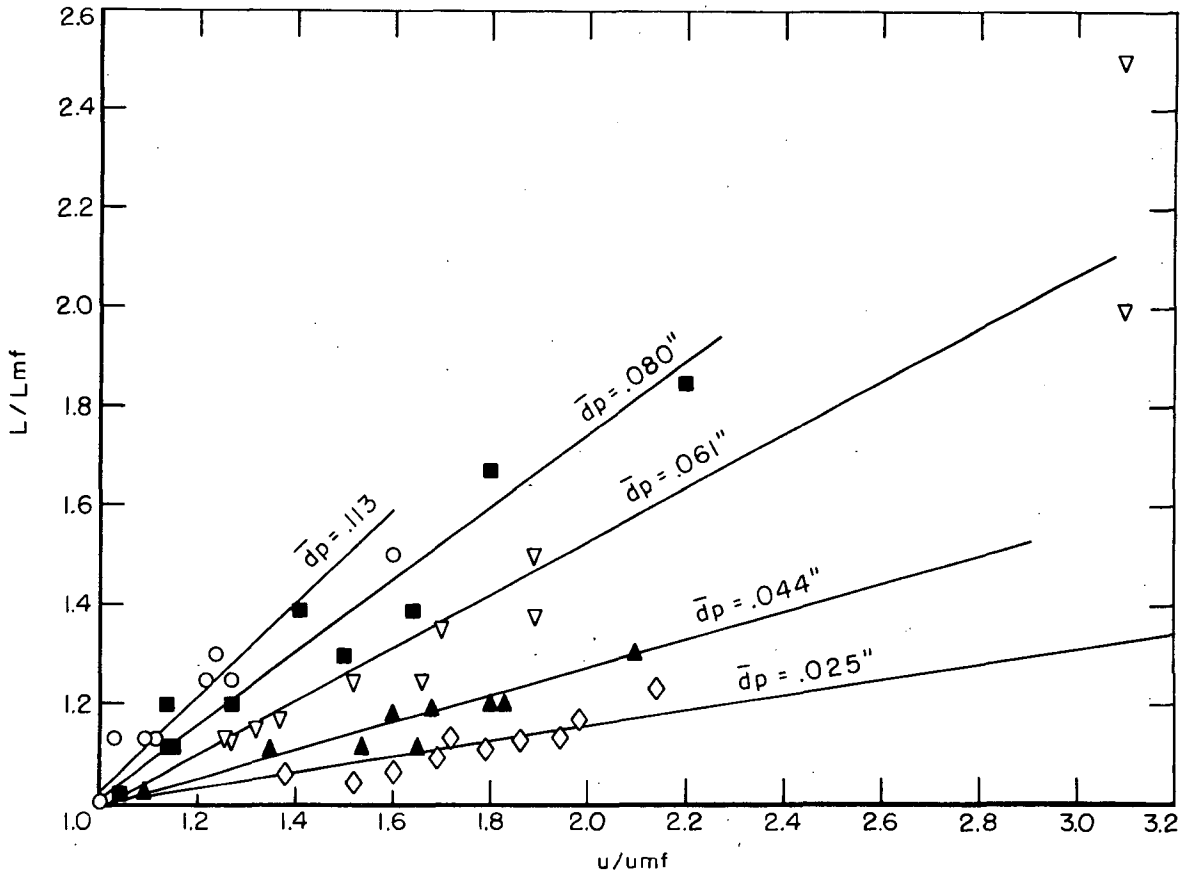

FIGURE 7a.- Effect of $\bar{d}p$ and u/U_{mf} on Bed Expansion.

L-12235

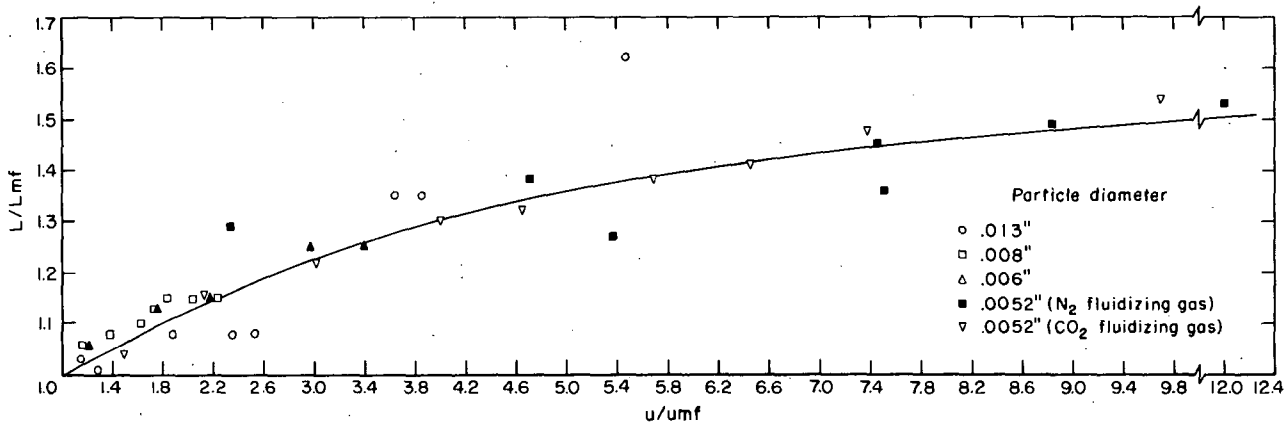

FIGURE 7b.- Effect of u/U_{mf} on Bed Expansion for $\bar{}_p$ <.025.

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