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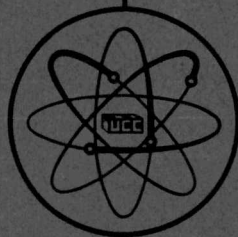
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Chemistry-General

DEVELOPMENT OF A CONTINUOUS COUNTERCURRENT
HIGH EFFICIENCY GAS-SOLIDS CONTACTOR

R. P. Levey, Jr.
A. de la Garza
S. C. Jacobs
H. M. Heidt
P. E. Trent



Y-12 Plant

UNION CARBIDE NUCLEAR COMPANY
DIVISION OF UNION CARBIDE CORPORATION

Oak Ridge, Tennessee

Acting Under U. S. Government Contract W7405 eng 26



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UNION CARBIDE NUCLEAR COMPANY
DIVISION OF UNION CARBIDE CORPORATION
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ABSTRACT

Experimental conversions of UO_3 to UO_2 and UO_2 to UF_4 in a batch fluidized bed pilot-scale contactor have demonstrated that conversion rates approaching those predicted from basic kinetic data are attainable. Further studies with fluidized beds in columns incorporating a very slight taper show that solids mixing with attendant product contamination is reduced by a factor of ten as a direct result of the taper.

Experiments with tapered fluidized beds made the design of continuous (as opposed to the step enrichment of a multistage contactor) countercurrent contactors practical. Design procedures, in generalized form, are presented. The procedures take into consideration heat transfer, mixing losses, reaction kinetics, and chemical equilibria.

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SUMMARY

Data obtained in batch conversions of UO_3 to UO_2 and UO_2 to UF_4 in a 5-inch diameter fluidized bed pilot plant are presented. These data indicate the pilot-scale conversion rates approximate laboratory thermo-gravimetric⁽¹⁾ rate measurements on the same materials. It is concluded then that pilot-scale conversion rates are limited only by the reaction kinetics. The pilot-scale conversion rates are noted to be several times greater than the rates observed in other fluidized beds processing similar materials.^(2,3) These higher rates are attributed to a more efficient state of fluidization and to the absence of losses in the batch tests due to intermixing of partially converted feed with the product stream.

Experiments in cylindrical and tapered glass columns indicate the taper greatly stabilizes the bed and makes the use of bed depths up to 14 column diameters practical. In a comparison of a tapered and a cylindrical column containing fluidized UO_3 , the mixing was found to be more than ten times as fast in the cylindrical unit.

The high efficiency and lack of bed mixing in tapered fluidized beds made the design of a countercurrent continuous contactor appear practical. A design procedure is outlined and an illustrative problem for converting UO_2 to UF_4 is presented. The performance of this design is compared to other UO_2 to UF_4 fluidized bed converters.

INTRODUCTION

The Y-12 Plant operated for the Atomic Energy Commission by the Union Carbide Nuclear Company has for years processed fission product free uranium. The uranium compounds are valuable because they contain a large fraction of U²³⁵. The nominal value ranges from ten to fifteen dollars per gram of contained uranium. In the past, most of this material has been processed manually in small batches. The manual technique served a dual purpose: inventory control and nuclear safety.

Any process improvements and expansions applied to enriched uranium processing must economically provide the capacity needed, nuclear safety, and absolute inventory control. In the case of the conversion of UO₃ to UF₄, a single batch-operated fluidized bed contactor was found to offer the most attractive over-all process. Experimental operation of a pilot plant proved completely successful with a 5-inch diameter unit, the largest permissible for nuclear safety. The bed depth in a fluidized bed of conventional design was found to be limited to approximately four diameters; thus the processing capacity of a 5-inch unit was established by this depth limitation.

The concept of a tapered column was first considered at that point in the development work. The use of such a configuration was demonstrated with bed depths up to 14 diameters and the fluidization was found to be more stable than in conventional beds of four diameter's depth. Thus, in batch operation using a tapered reactor, the capacity of a unit could be increased by a factor of approximately three (actually 14/4). The development work with cylindrical and tapered columns from June 1957, to January 1958, is described in this report.

EXPERIMENTAL

BATCH CONVERSION OF UO_3 TO UF_4 IN A FIVE-INCH DIAMETER FLUID BED UNIT

The pilot plant unit is shown schematically in Figures 1 and 2, Appendix A. Referring to Figure 1, reagent gas is preheated in a resistance heater. This heater consists of a length of electrically isolated pipe. Heating is obtained by applying a low voltage directly to the pipe. The heat load is approximately 10 KW. Good compliance to the demand is obtained by minimizing the heat capacity of the preheater.

The bed support and gas disengager, shown schematically in Figure 1, provide absolute inventory control since they are permeable only to the gas. This development is described in a separate report.⁽⁴⁾ The entire system is fabricated of Inconel because of demonstrated corrosion resistance.

Temperature control for heating the reactor is conventional and automatic. During cooling, the temperature is controlled manually with a cooling coil welded to the vessel. The temperature control must be maintained within $\pm 20^\circ$ F during reduction to preserve product reactivity, and air was found to be the best cooling medium for this purpose. The peak heat load during the hydrofluorination is several times that of the peak reduction heat load but the required control range is approximately $\pm 100^\circ$ F because this material is less temperature sensitive. Water is preferred for cooling during this cycle.

Typical Conversion Runs

The pilot unit described was used in a campaign to assess the longer term operating problems that might arise. During a 112-hour test period only four hours' downtime was required for routine maintenance. The UO_3 feed used was 20 to 40 mesh material made by the continuous calcination of uranyl nitrate at Hanford, Washington.⁽⁵⁾ Feed analyses are given in Table I, Appendix A. The average contamination pickup during conversion was 44 ppm of iron, less than one ppm of copper, 24 ppm of chromium, and 46 ppm of nickel. The conversion cycle, including cooling time, loading, and unloading, was less than six hours. The reduction cycle was one hour at $1,050^\circ$ F. The hydrofluorination cycle was one hour at 400 to 600° F and one hour at 600 to $1,300^\circ$ F. As shown in Table II, reduction averaged 99.3 percent efficiency and the hydrofluorination averaged 98.8 percent efficiency.

Fluidization Velocity

The velocity required for good fluidization of the reacting materials was found to be dependent upon the material size, fluidizing medium, pressure, and temperature. The velocity used in experimental conversions and referred to as the "fluidization" velocity was that required to produce a bed pressure drop equivalent to the bed weight. This velocity was experimentally determined with prevailing reaction conditions for each new feed material. Conversion experiments with powders ranging from 20 to 40 mesh to 100 percent finer than 200 mesh were performed. The velocities required were from 0.1 to 3.5 ft/sec.

The experimental data are shown in Table III and graphically in Figure 3. The solids carry-over was from two to five percent with a freeboard above the expanded bed of approximately 2-1/2 diameters.

HEAT TRANSFER MEASUREMENTS

During both reduction and hydrofluorination operations, many temperature traverses of the bed were taken, both transverse and longitudinal. No variations greater than 2 or 3° F were found within the bed when it was operating properly. Bed temperature measurements were never made closer than 1/2 inch from the vessel wall. The reactor skin temperatures in conjunction with the temperatures one-eighth to one-fourth inch away from the outside vessel skin made possible rough estimates of the over-all heat transfer.

Two types of heat transfer measurements are possible, during reaction and during heating or cooling. The measurements taken during reaction were adjusted for the fractional conversion which was estimated from the reagent added. The measurements taken during heating or cooling were based upon known heat capacities of the bed. Neither type of analysis were accurate because a steady state was not attained. During the experiments, the gas entered the reactor at bed temperature so that no adjustment for sensible heat in the gas was necessary. The latter type of measurement was probably more reliable as indicated by the results shown in Table IV. Over-all heat transfer coefficients equal to those listed as "powder to wall" could be realized using a water or air jacket cooling system. The measured values, 50 to 60 Btu/hr-ft² - °F, were somewhat larger than computed estimates 22 to 32 Btu/hr-ft² - °F. (6)

PNEUMATIC SOLIDS TRANSFER

The batch fluid bed conversion studies previously described were made by manually loading and unloading the reactor. The procedure along with appropriate weighings positively identified the solids inventory at all times. The labor saving convenience of pneumatic solids feed and discharge was investigated. Tests with both pressure and vacuum transfers were performed with 20 to 40 mesh solids as well as solids 100 percent finer than 325 mesh. The pressure and vacuum transfers were successfully made with UF₄, UO₂, and UO₃.

The experimental system shown, Figure 4, was composed of a feed and product receiver and the fluid bed conversion unit previously described. The units were connected through a 200-foot length of 3/4-inch tubing containing ten 180-degree, three-inch radius bends. This tortuous path was employed to explore any plugging tendencies the transported powders might show. The standard test procedure was to charge 40 pounds of UO₃ to the feed hopper and pneumatically transport it to the reactor. The UO₃ was converted to UF₄ in the normal way and the converted inventory was returned to the product hopper.

All pressure transfers were performed at 20 to 40 ft/sec superficial air velocity. The vacuum transfers were performed at approximately one-half atmosphere with approximately 40 to 80 ft/sec superficial air velocities. Transfers were made with 40- to 140-pound solids per pound of motivating gas. A summary of the transfer data is shown in Table V.

STUDIES WITH DEEP BEDS

In fluidizing sized spheroidal UO_3 particles in cylindrical glass columns, it was noted that boiling was more violent at the top than at the bottom. When a static bed deeper than four diameters was fluidized, large bubbles appeared near the top before the bottom could be fluidized. As deeper beds were employed, violent eruptions with consequent inefficient contacting resulted. A typical deep bed at rest and expanded is shown in the photograph in Figure 5. For example, a 20 to 40 mesh UO_3 bed ten diameters deep must expand 25 percent before complete fluidization is attained. The same diameter bed only four diameters deep need expand only 2 percent for complete fluidization as shown in Figure 6. In the UO_3 to UF_4 conversion, the practical operating range in bed expansions was less than 5 percent. In a bed four diameters deep an expansion of 2 percent was required to obtain fluidization throughout. Thus, it was shown qualitatively that cylindrical beds of perhaps five diameters' depth represent the largest practical operating inventory. For UO_3 in a 5-inch diameter reactor, this was approximately a 60-pound charge.

The capacity of a batch system employing cylindrical vessels can be increased only by use of multiple reactors. Research on methods of operating a higher capacity small diameter vessel was undertaken. In studying the fluidization process in glass cylinders, it was often noticed that the upper surface fluidized first and that this process proceeded downward through the bed until the particles adjacent to the distribution plate were fluidized. By probing the bed manually with a vertical rod this observation was proven correct as the rod could be dropped (or forced) only through the bubbling portions of the bed. It was reasoned that the pressure drop resulting from the fluidization process, approximately 1.75 psi/ft for UO_3 , must be accompanied by an equivalent velocity increase. This velocity change per unit height then might be the basic cause of the greater bed expansion near the upper surface. If this were true, a tapered vessel might be fabricated which would fluidize first on the bottom and last on the top.

A glass vessel 3-inch diameter x 6-inch diameter x 36 inches long was constructed with a taper of approximately 2.5 degrees. The fluidization of UO_3 in this vessel occurred first at the bottom and was quite violent before the upper surface was visibly disturbed.

In view of this finding, an optimum angle based on the pressure drop-velocity concept could be defined. The following relationship was found to approximate the angle.

$$\tan \phi = \frac{D_t}{2L} \left[1 - \left(\frac{1}{1 + \frac{L \Delta p}{P_t}} \right)^{1/2} \right] \quad (1)$$

A table of nomenclature is shown on page 20.

The bottom diameter is approximated by

$$D_b = D_t \left[\frac{1}{1 + \frac{L \Delta \rho}{P_t}} \right]^{1/2} \quad (2)$$

A glass vessel of approximately this specification (assuming $p_t = 14.7$, $\Delta p = 1.7$ psi/ft) was fabricated. The actual dimensions were 3-inch D_b x 4-inch D_t x 40-inch L over-all. Fluidization of UO_3 in this unit was visually smooth and uniform throughout. The rod test described and bed pressure drop proved the only positive way to identify the fluidized state since no gas bubbles were seen. This stabilization of a fluidized bed by a tapered vessel has been observed by Omae(7) in liquid-solid systems. Height-to-diameter ratio $\left(\frac{L}{D_b}\right)$ for this vessel was 12:1 and the angle of taper was 0.88 degree.

A third column with the same taper but six feet deep was fabricated of glass.

This unit was 5-inch D_b x 6-inch D_t x 72-inch L , $\left(\frac{L}{D_b}\right)$ was greater than 14:1.

Observations of fluidization of UO_3 with this unit established that a 6-foot deep bed was thoroughly fluidized, as indicated both by pressure drop and the rod test at outlet velocities approximately 75 percent of that required in a cylindrical vessel of the same diameter. The bed expansion was approximately 0.7 percent in six feet. A photograph of this bed at rest and fluidized is shown in Figure 7.

Observations of tapered column in action established that:

- a. The mixing pattern within the bed was different than in a cylindrical unit of similar proportions.
- b. Bed expansion in the tapered units was only 10 percent of the expansion in a cylindrical unit of similar proportions. Furthermore, the expansion per unit length appeared to be linear in the tapered unit (i.e., $\left(\frac{de}{dL} = k\right)$ while in the cylindrical units, expansion was proportional to the height $\left(\frac{de}{dL} = kL\right)$.

CONVERSION OF UO_2 TO UF_4 IN A TAPERED FLUIDIZED BED BATCH REACTOR

A reactor of Inconel was fabricated which was 3-inch D_b x 6-inch D_t x 39-inch L , which was an angle of about two degrees. This taper was probably greater than the optimum. Five conversion runs were made with various types of powders. Fine beds were used since these tend to cake and provide a more severe test. In no case was any difficulty encountered. Fluidization velocities the same as employed with the cylindrical reactor were used. The results are shown in Table VI.

DETERMINATION OF A MIXING COEFFICIENT FOR THE MIXING OF TWO FLUIDIZED SOLIDS

In a fluidized (cylindrical) bed, the boiling tends to maintain thorough mixing of inventory. In a continuous contactor involving feed and product streams this mixing causes short circuiting of feed, or partially converted feed into the product stream. The productivity can be estimated only if the distribution of residence times and the conversion efficiency as a function of residence time is known. The purpose of the experiments described was to study the mixing of UO_3 , UO_2 , and UF_4 in fluidized beds in the absence of reaction. The experiments were performed both in cylindrical and tapered columns to maintain valid comparisons.

In the first series of experiments a column was loaded with equal volumes of weighed, analyzed, and screened constituents. The bed was fluidized and samples were withdrawn from one end at fixed time intervals for chemical analysis. In experiments with equal layers, great difficulty arose in initiating fluidization because the interface between layers acted as a barrier, i.e., the layers acted as though they were immiscible for a short time. It was concluded that the addition of a very small amount of one component to a bed already fluidized would eliminate this problem. A new problem arose, however, in that the chemical analyses were not sufficiently reproducible. This was because of the very low concentrations of additive involved and because dusting of the additive tended to poison the system. The results of these experiments, though not satisfactorily reproducible, indicated that the mixing in a cylindrical bed was 20 to 50 times faster than in a tapered bed. The numerical results of the experiments are summarized in Table VII. The development of numerical analysis is shown in Appendix B.

Actually, none of the experiments performed reproduced the specific circumstances of interest; namely, a continuously fed and discharged countercurrent unit. An experiment was planned in which a fluidized bed of UO_3 was continuously fed with UO_3 which was partially reduced so that the UO_2 content was approximately 10 percent. This procedure permitted the use of components of nearly identical physical properties. Two experiments were performed under identical conditions of fluidization and feed rate in a cylindrical and in a tapered column. The columns chosen were a 5-inch D x 5-foot L cylinder, and a 5-inch D_b x 6-inch D_t x 5-foot L tapered column. The units were fed so that the weight fed was equal to the inventory weight after 3.14 hours. The total material removed in five-minute intervals was quartered to obtain duplicate samples for analysis. The experimental set-up is shown schematically in Figure 8. Results can be considered in terms of the concentration of feed in product at any given time (or fraction of turnover time). For example, if no mixing occurred, no feed would appear in the product until turnover time at which point the concentration would instantaneously become 100 percent, i.e., a square wave (piston flow) would result. If, on the other hand, there existed no concentration gradient in the bed (instantaneous mixing), then feed would at all times be found in the product from beginning to end. Experimental results are tabulated in Table VIII. The graphic comparison shown in Figure 9 demonstrates, for example, that if a fluid bed reaction were to be carried out which was thermogravimetrically 99 percent complete in one hour, 98 percent conversion would be realized in the tapered unit, 81 percent in a cylindrical

unit, and 74 percent in a totally mixed unit. The computation of the mixing coefficient "M" for the two columns has been approximated. The values are shown:

Cylindrical Column

3.98 ft²/hr

Tapered Column

0.4 ft²/hr

DISCUSSION

FLUIDIZATION EXPERIMENTS

As gas is passed up through a column of particulate solids, the pressure drop will increase as the flow is increased. In a bed less than four diameters deep, as shown in Figure 10, the surface appears to boil first. When a flow is reached at which a rod can be freely passed, the pressure drop is approximately equal to the bed weight. This point is shown in Figures 5 and 10. Further gas flow increase causes more violent boiling and a peak pressure drop can be attained. Additional flow increases bring about a slight pressure decrease, although the pressure drop does not fall below a pressure drop equivalent to the bed weight. The fluidized state, in this text, refers to beds through which a rod may be freely passed and in which the entire bed weight is manifest as pressure drop.

BATCH CONVERSION EXPERIMENTS

Pilot plant conversion UO₃ to UO₂ and of UO₂ to UF₄ is approximately 99 percent complete in three hours' total reaction time as shown in Table II. It also was observed, see Figure 11 and 11a, that this conversion can be carried out in a thermo-balance at about the same rate. It is concluded that under the pilot plant conditions reaction kinetics is the rate controlling step in the conversion process. In these batch conversions the following processes can be rate controlling phenomena:

- a. Heat transfer.
- b. Reagent delivery.
- c. Chemical equilibrium.

The heat transfer and chemical equilibria will be discussed separately.

The reagent velocity in many cases controls the retention time required. In such fluid bed conversion units, reagent velocities of less than one-half foot per second are used because the size of the material is <100 mesh. Fluidization of materials from 20 to finer than 325 mesh was investigated and has been studied by others.⁽⁸⁾ Materials finer than 200 mesh are extremely difficult to fluidize because of channeling problems. No work with particles

larger than 20 mesh is reported. The reagent consumption rate in a fluidized bed reaction is governed by the chemical activity of the particle while the over-all gas velocity is generally governed by the particle size. With a feed of given activity the time required to perform the batch conversion to UF_4 was found to vary from 2 - 15 hours when the particle size varied from 20 to 200 mesh as shown in Table III. Thus the reagent velocity becomes the rate limiting process in conversion unless sufficiently large particles are used. Consider a 60-pound UO_2 charge in a 5-inch reactor, and assume the desired conversion is kinetically possible in two hours and that this is to be the rate controlling process. As shown in Figure 3, particles greater than 65 mesh must be employed if this two-hour conversion is to be achieved. In practical cases, even larger sizes can be dealt with. The percent conversion of various sizes of UO_3 powders has been studied with a thermobalance, and sample data are shown in Table IX and plotted in Figure 11. As seen from the plots, the gain in conversion does not compensate for the loss in time required to supply the stoichiometric HF in a fluid bed contactor. No numerical analysis of the optimum particle size-reactivity relationship for a given design problem will be made since for the case of interest, $UO_3 \rightarrow UF_4$, powders larger than 60 mesh are clearly preferable.

HEAT TRANSFER EXPERIMENTS

Experimental heat transfer measurements are found to be independent of fluidization velocity. The range of temperature differences over which these data were obtained was 100 to 300° F. In the batch reactor from which the data shown in Table IV were obtained, the heat release per unit time is extremely nonuniform. This results from the shape of the conversion-time relationship shown in Figure 11. The cooling area required, however, to handle a load of this kind can be computed. Actually, in designing a continuous system employing a tapered column, the heat release would be more uniformly distributed along the vessel.

MIXING EXPERIMENTS

The absence of bubbling in tapered columns, as well as their almost linear expansion per unit height, indicated a basically different type of fluidization might exist in tapered columns from that in cylindrical columns. The utility of the tapered design would depend upon studies of whether or not particle movement was sufficient to prevent caking in conversions where this is normally a problem. The successful conversion of fine UO_2 powders to UF_4 in a tapered column (see Table VI) indicated the tapered column is no more susceptible to caking than is a cylindrical unit. The intermixing of inventory was shown (Table VII) to be only a fraction as much in tapered columns as in cylindrical columns. In addition, the stability of deep beds (up to 14 diameters) was demonstrated. In view of these findings, it appeared likely that a countercurrent contactor of high efficiency for both solids and gas could be designed utilizing a single tapered contactor. The general problem of designing such a unit to carry out a conversion involving a first order reaction in the presence of mixing losses is taken up in Appendix C. The presence of a first order reaction was assumed because both the reduction and hydrofluorination reactions closely approximate this after the first 5 to 10 minutes. This point is brought out

in Figure 11. The mixing in fluidized bed systems has been studied by other workers(9 thru 14) for both phases during mass transfer operations. The problem of mixing in the presence of a first order reaction has also been considered.(9) However, in all cases observed mixing was so rapid that when high conversions or slow reactions are encountered, multiple staged reactors have been the only solution to the problem. The rapid mixing in cylindrical column experiments, as seen in Table VII, clarifies this point. The need for staging reactors in UO_2 and UF_4 production(2,3) illustrates this point. However, without the numerical analysis it is clear that a five-foot tapered column which is a single stage (Table VI, Figure 9) could produce greater than 99 percent product with a one-hour retention providing the reaction kinetics was the rate controlling process. This indicates the practicality of carrying out slow reactions or obtaining equivalent multistage results in a single envelope. The use of tapered columns could provide the advantages of multistage contacting without the disadvantage of multistage units such as duplicate feeds, controls, distributors, disengagers, etc.

Based on the experimental work described in Appendix B, a mixing coefficient "M" (diffusivity) under conditions of normal fluidization for 20 to 40 mesh UO_2 , UO_3 and UF_4 was found to be $< 0.4 \text{ ft}^2\text{-hr}^{-1}$. The reactor length required by mixing and kinetic considerations is given by

$$1 - c = \frac{4H}{\left[(H + 1)^2 e^{\left(\frac{H - 1}{2m}\right)} \right] - \left[(H + 1)^2 e^{-\left(\frac{H + 1}{2m}\right)} \right]} \quad (3)$$

Where

$$m = M/uL$$

$$H = (1 + 4 m R)^{1/2}$$

$$R = \frac{kL}{u}$$

The length called for by equation (3) assumes that mixing and reaction proceed independently. This assumption has not been experimentally verified. This reactor length is not necessarily sufficient to produce product concentration since it assumes kinetics and mixing control conversion.

In developing the conversion on the basis of the reaction kinetics and a constant mixing coefficient, two other cases of interest are shown:

If there is no mixing ($m = 0$, "piston flow")

$$(1 - c) = e^{-\frac{kL}{u}} \quad (4)$$

If there is total mixing ($m \rightarrow \infty$), no gradient, the entire bed is homogeneous

$$(1 - c) = \frac{1}{1 + \frac{kL}{u}} \quad (5)$$

In the experimental data of Figure 9, the results of all three cases are plotted. The experimental lines represent mixing coefficients of 4.0 and 0.4 ft²/hr, cylindrical and tapered units, respectively, while the left and right lines show the results of equations (5) and (4), respectively. The mixing in cylindrical columns has been shown to approach the total mixing case while that in tapered columns can be compared with the case for piston flow. A comparison of the column length required for given conversion is tabulated below:

$$c = 0.99$$

$$k = 1.0 \text{ hr}^{-1}$$

$$u = 2.0 \text{ ft/hr}$$

$$M = 0.4$$

	<u>No</u> <u>Mixing</u>	<u>Tapered</u> <u>Column</u>	<u>Complete</u> <u>Mixing</u>
Required Length:	9.2 ft	11 ft	198 ft

Thus, the performance of a tapered column when considered in terms of relative length is shown to approach the ideal case, while the need for staging ordinary cylindrical units is also illustrated.

EFFECT OF CHEMICAL EQUILIBRIA

The discussion of the influence of mixing on reactor design, especially for tapered units, is based on the assumption that in a continuous unit the kinetics would be the rate controlling phenomena as was observed in the batch experiments. Studies with a five stage UO₂ to UF₄ conversion unit indicated that near (approximately 90 percent) equilibrium values of exit HF can be attained. This was observed only when a deficiency of HF was employed and poor UF₄ conversion resulted. Thus, the influence of the chemical equilibrium on reactor size must be considered when both conversion and reagent usage greater than 90 percent are desired. This problem has been treated for the conversion of UO₂ to UF₄ by assuming the transport of HF from bulk gas to bulk solid is proportional to a driving force ($y - y^*$) such that

$$J = k (y - y^*) \quad (6)$$

This rate equation can be combined with material balance information to obtain:

$$LS = \frac{4F}{r\rho k} \int_{x_b}^{x_t} \frac{dx}{x (y - y^*)} = V \quad (7)$$

Equation (7) can be integrated to determine a required column volume when conditions of conversion and HF usage, temperatures, and feed rates are specified. The numerical analysis of the problem is given in Appendix D. This analytical method is then utilized, Appendix E, to estimate a contactor volume required for producing 1,000 pounds per hour of UF_4 with 99 percent conversion and 95 percent usage of HF.

The equilibrium partial pressure of HF for the reaction at various temperatures has been studied.⁽¹²⁾ The equilibrium partial pressure is plotted over a range of temperature as shown in Figure 12. The equilibrium constants from which these data were computed are plotted in Figure 13. The need for operating at low temperatures to get good HF usage is illustrated in that at $1,000^\circ$ F the required partial pressure of HF must be 48 percent while at 400° F it need be but 0.4 percent to obtain conversion. On the other hand, the conversion rate constant rises with temperature so that inventory can be reduced or conversion improved by high temperature operation. Thus, the need for designing a reactor with zoned temperature is qualitatively shown. The optimum temperature gradient has been studied by others⁽²⁾ for UF_4 production and found to be 750° to $1,100^\circ$ F in five stages. The material used was relatively inactive and had an equivalent first order rate constant of approximately 1.0 hr^{-1} at $1,000^\circ$ F. In experiments in the 5-inch diameter batch reactor, a temperature cycle of one hour at $400\text{-}600^\circ$ F and one hour at $600\text{-}1,200^\circ$ F gave a minimum time for 99 percent conversion. Reagent excess of 20 percent was used.

DESIGN OF A TAPERED FLUID BED

A procedure for designing a large scale production unit for converting UO_3 to UF_4 must be developed in order to estimate the performance and dimensions of such a converter. These estimates can then be compared to performance and dimensions of currently reported UO_3 to UF_4 conversion systems. The design of the tapered unit presented is based on the material presented in this report.

Controls and Instrumentation

The main control problems in a large scale design are solids and reagent flow-temperature control and fluidization control. The methods proposed are described and shown schematically in Figures 14 through 18.

Solids Flow - The feed rate can be set by set point on FRC (flow recorder controller) which operates a calibrated screw feeder. The control system is shown schematically in Figures 14 and 18.

Product flow is maintained to keep a constant reacting inventory. The product is pneumatically conveyed by airlift. The lift is actuated through a controller which senses the column bed pressure differential. Thus, the bed level can be set as desired. This is shown schematically in Figure 15.

Series Operation - In cases where two reactions must be carried out, as for example the reduction and hydrofluorination of UO_3 to produce UF_4 , two contactors are required. For this case, only one feeder would be used and it

would control the over-all product rate. Two airlifts would be required. The first lift would discharge directly into the disengaging chamber of the second reactor so that only one product cyclone would be required for series operations.

Temperature Control - The conversion of UO_2 to UF_4 would be carried out at two temperatures. The heat is supplied in three zones, as shown in Figure 16. The low temperature is required to obtain good reagent usage while the high temperature is required to obtain final product level in a short exposure. The zone control is simply "on-off" conventional resistance heat.

The preheater operation is based on supplying low voltage-high current power to a selected length of feed gas pipe. The pipe is isolated from ground on the upstream end (see Figure 16) and the reactor itself is grounded. This has been found to assure delivery of gas at the desired temperature by providing continuity of heat. The power is varied through a voltage regulator so that modulation control is available. A transmitter for converting thermocouple output to an air output (E to A) is employed for this purpose.

Cooling control is gained in a manner similar to the preheat control. Cooling steam is throttled through a transducer which converts a thermocouple signal to an air signal.

Fluidization Control - The bed fluidization can be controlled on the basis of bed pressure drop. However, since the best operating condition is always in a region of small change in drop for large changes in gas flow rate, see Figure 10, this method of control is difficult in practice. Fluidization at any column condition (temperature, gas, pressure) can be controlled on the basis of velocity if the velocity at one condition is known. This velocity is given by

$$v = \frac{\rho}{4.5} (1 + 0.0022 t)^{-1} v_1 \left(\frac{\mu_1}{\mu_2} \right)^{.23} \quad (8)$$

for UO_3 in nitrogen at 100° F.

Where

v_1 = fluidization velocity of UO_3 in 100° F nitrogen, ft/sec

μ_1, μ_2 = viscosities of nitrogen and reactor gas in consistent units

This method of control has been found impractical not only because of variations in pressure, gas, temperature, etc., but because automatic compensation for these variables requires analog type equipment.

Control by the rod method, shown schematically in Figure 17, is based on the observation (discussed earlier in this report) that the resistance to movement in a static bed is large while in a fluidized bed it is very slight. Referring to Figure 17, the sensing rod is excited at its resonant frequency by the oscillator shown. The vibration amplitude of the rod is amplified by the pick-up shown.

This amplitude meter feeds the conventional electric to air converter which controls the reagent gas flow. The rod response depends basically upon bed fluidity. The amplitude output is quite sensitive to bed expansion as shown in Figure 18.

Column Dimensions

Column dimensions are developed on the basis of feed and product compositions required. The minimum operating inventory for design production is the basic criteria. The procedure is presented in sufficiently detailed manner as to permit its use in the design of units other than for the manufacture of UF₄. The reactor size controlling processes are listed:

<u>Process</u>	<u>Dimensions</u>
Cooling Reaction	$D L = \frac{3 q w}{\pi h \Delta t}$
Reagent Supply	$D^2 = \frac{(1.1)(4)(f)}{\pi v (1 - \epsilon)}$
Mixing:	
Case 1, Piston Flow	$D^2 L = \frac{\ln \left(\frac{1}{1 - c} \right)}{\frac{\pi \rho k}{4 w}}$
Case 2, Complete Mixing	$D^2 L = \frac{4w \left(\frac{c}{1 - c} \right)}{\pi \rho k}$
Case 3, Constant Limited	See Figures 19 and 20 and Appendix B
Reagent Efficiency	$D^2 L = \frac{4F}{r \rho k} \int_{x_b}^{x_t} \frac{dx}{x (y - y^*)}$

In a given design problem all the dimension groups can be numerically represented. The two basic volume determining groups result from the mixing process and the reagent gas stripping process. The larger of the two represents the minimum operating volume (process inventory). There is a minimum column diameter resulting from the reagent volume that must be passed while the reagent velocity is limited by requirements for good fluidization. The factor 1.1 (see tabulation above) is included to provide sufficient inert gas flow to maintain fluidization control. The column area may be fixed by cooling requirements. Since approximately 90 percent of the UO₂ conversion occurs in the upper third of the column, a factor 3 (see tabulation above) is included in this criteria. From the four criteria listed, a length and diameter for minimum inventory can be computed.

Use of the equilibrium data together with the coefficient developed in equation (6), a column volume required for given reagent usage and conversion can be computed. A temperature or temperature gradient must be assumed.

Since a minimum volume is not implied for the assumed temperatures, several computations would be required to develop a truly minimum inventory. Experimentally, the batch conversion results showed that 400° F was the lower operating limit and that above 1,300° F caking occurs. The minimum time for batch conversions was obtained when 80 to 90 percent conversion was obtained at 400 - 500° F. The remainder was converted while heating to 1,200° F. In the example design problem shown in Appendix E, zone temperatures of 400° and 1,050° F with conversions to 90 and 99 percent, respectively, were assumed. The inventory of Appendix E is minimum only for the assumed gradient and other gradients would have to be computed to arrive at truly minimum dimensions for the process load.

COMPARATIVE PERFORMANCE OF TAPERED COLUMNS

The illustrative design problem shown in Appendix E together with the discussion on the design of fluidized bed contactors provides a sound procedure for practical cases of interest. The procedure is summarized in Table XI. As shown in Table XI, a contactor of minimum inventory can be designed for any conditions in conjunction with experimentally determined constants. The size controlling phenomena (whether mixing, cooling, HF usage, etc.) is determined and considered in the choice of dimensions.

The conversion of UO_2 to UF_4 has been carried on by others^(2,3) in fluid bed contactors. In each of these cases a tapered unit was sized, as shown in Appendix E, to process the identical material to the same rate and purity or conversion. The estimates are summarized in Table X. The advantages of the tapered reactor design are seen in terms of increases in productivity of 150 to 650 percent.

TABLE OF NOMENCLATURE

D	=	column diameter, ft
F	=	feed rate of solids reactant, lb mol/hr
G	=	total molar gas flow, lb mols/hr
I	=	inert gas flow lb mols/hr
J	=	transport of reactant gas (HF), lb mols/hr-ft ² - (y - y*)
J*	=	transport of reactant gas (HF), lb mols/hr
L	=	active bed depth of column length, ft
M	=	mixing coefficient for fluidized solids (diffusivity), ft ² /hr
R	=	fluidized bed superficial surface area, ft ² /ft ³ column
S	=	column cross sectional area, ft ²
V	=	column volume, ft ³
c	=	fractional conversion of feed to product, dimensionless
e	=	fractional bed expansion from rest to fluidized state, dimensionless
f	=	reagent gas flow, outlet conditions, to supply stoichiometric requirement, ft ³ /sec
h	=	heat transfer coefficient, powder to vessel wall, BTU/hr-ft ² -°F
k	=	equivalent first order reaction rate constant, hr ⁻¹ , result of thermogravimetric measurements
p	=	partial pressure of reagent gas, psia
Δp	=	bed pressure drop, psi/ft bed
q	=	heat release due to reaction, BTU/lb feed
r	=	superficial surface area of reactant, ft ² /lb mol
t	=	temperature, °F
u	=	superficial solids velocity, ft/hr
v	=	superficial gas velocity for fluidization, ft/sec

w = solids feed rate, lb/hr

x = mol fraction uranium, solids

y = mol fraction reactant, gas

t, b = subscripts (indicate top and bottom of fluidized bed)

ϵ = bed void volume at rest, dimensionless

ρ = bed density in action, lb/ft³

μ = gas viscosity, consistent units

ϕ = column taper measured as the angle included between the column wall and vertical line

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APPENDIX A

TABLES AND FIGURES

TABLE I
ANALYSIS OF UO_3 FEED FOR BATCH PILOT TESTS

Item	Unit	Result
Total U	g U/g	.82550 \pm .0001
Total U ⁺⁶	g U/g	.82550 \pm .0001
Total U ⁺⁴	g U/g	0
Screen Analysis	Tyler Mesh	-20 +35
Density, Tap	g/cc	4.17
Rate Constants for Conversion (1st order):		
Reduction	hr ⁻¹ at 1,050 ^o F	7.0
Hydrofluorination	hr ⁻¹ at 1,000 ^o F	2.3

TABLE II
SUMMARY OF PILOT PLANT BATCH CONVERSION RESULTS
IN A 5-INCH DIAMETER CYLINDRICAL REACTOR

Run No.	UO_3 Charged (lb)	UF_4 Tap Density (g/cc)	Reduction (%)	UF_4 (%)
46	40	3.5	98.5	99.2
47	40	3.4	99.0	99.9
48	40	3.3	98.7	99.9
49	40	3.5	99.6	100.0
50	40	3.6	97.4	100.6
51	40	3.6	99.6	99.8
52	40	3.5	99.9	99.7
53	40	3.6	99.8	98.1
54	40	3.8	99.4	89.8
55	40	3.5	99.0	101.2
56	40	3.6	99.6	98.1
57	40	3.6	99.6	98.5
58	40	3.5	99.5	100.9
59	40	3.4	98.7	98.2
60	40	3.6	100.0	98.5
61	40	3.4	100.0	98.5
62	40	3.4	99.1	98.4
Average		3.5	99.3	98.8
Reduction Time:		1 hr at 1,025 ^o - 1,050 ^o F		
Hydrofluorination Time :		1 hr at 400 ^o - 600 ^o F 1 hr at 600 ^o - 1,300 ^o F		

TABLE III
SUMMARY OF FLUIDIZATION VELOCITY DATA

Series Feed Size (Tyler)	Gas Velocity For Fluidization (ft/sec)	Hydrofluorination Time for > 98% Conversion (hr)
-20 +35	3.0 - 3.5	2
-35 +65	1.3 - 1.5	2.5
-65 +100	0.6 - 0.8	4
-100	0.3 - 0.4	5
-200 (50% -325)	0.1 - 0.2	15

TABLE IV
HEAT TRANSFER COEFFICIENTS OBTAINED DURING SELECTED
HYDROFLUORINATION RUNS

No.	Feed Particle Size (Tyler)	Powder to Wall (Btu/hr-ft ² -°F)	Wall to Air (Btu/hr-ft ² -°F)
20	20 - 40	49	6
21	40 - 60	63	13
22	20 - 40	58	6
36	20 - 40	65	19
<u>Selected Runs Based on Heating and Cooling Cycles</u>			
20	20 - 40	60	
21	40 - 60	51	
36	20 - 40	62	

TABLE V
SUMMARY OF PNEUMATIC TRANSFER RESULTS

Material Transferred	Material Size (Tyler Mesh)	Temperature of Solids (°F)	Transfer Distance (ft)	Pressure Transfer Lb Solids Lb Motivating Fluid	"Heel" Remaining in System (%)	Vacuum Transfer Lb Solids Lb Motivating Fluid
UF ₄	20 - 40	200	15	80	17	133
UO ₃	20 - 40	70	200	67	18	45
UF ₄	20 - 40	1,000	200	67	9	40
UF ₄	-325	70	200	30	--	--

TABLE VI
A SUMMARY OF CONVERSION RUNS MADE IN A
TAPERED FLUIDIZED BED BATCH REACTOR

Run No.	UO ₃ Charged (lb)	Mesh Size (Tyler)	Reduction Time (hr)	Hydrofluorination (hr)	Reduction (%)	UF ₄ (%)
70	20	-200	6	--	90.4	Not Hydrofluorinated
71	20	-200	6	8	99.6	97.9
72	20	-200	4	6	97.0	86.8
Special	20	20-40	2	4	99.0	96.0

TABLE VII
SUMMARY OF MIXING EXPERIMENTS

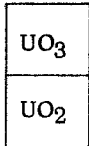
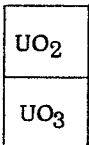
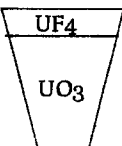
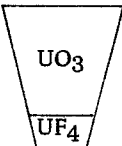
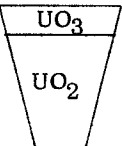
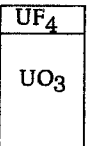
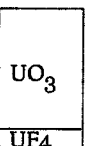
Experiment No.	Setup	Diameter (in)	Length (in)	Equilibrium Time (hr)	Mixing Coefficient ft^2/hr
1		4	15	0.03	10.0
3		4	18	0.02	20.0
7		4-5	72	24.0	0.3
8		4-5	48	24.0	0.1
9		4-5	76	24.0	0.4
10		5	49	1.5	7.0
11		5	49	1.0	10.0

TABLE VIII
 A SUMMARY OF RESULTS OBTAINED IN THE CONTINUOUS
 COMPARATIVE MIXING EXPERIMENT

Elapsed Time (hr)	Turnover Time (%)	Feed in Product Taper (%)	Feed in Product Cylinder (%)	Feed in Product Instantaneous Mixing (%)
0.1	3.2	0.1	No Sample	(Computed)
0.2	6.4	0.2	1.5	5.0
0.4	12.7	0	2.0	9.8
0.5	15.9	0	5.4	14.3
0.7	22.3	0	11.1	18.5
0.8	25.5	0	13.7	22.6
1.0	31.8	0.2	17.9	26.4
1.2	38.2	0.8	25.3	30.1
1.4	44.6	1.2	26.1	33.5
1.5	47.7	1.6	Broken	36.7
1.7	54.1	1.9	29.4	39.8
1.8	57.3	3.6	36.4	42.7
2.0	63.7	8.8	34.0	45.4
2.2	70.0	21.3	45.8	48.1
2.4	76.5	27.3	43.8	50.6
2.5	79.5	46.6	45.7	52.9
2.7	86.0	55.8	50.5	55.2
2.8	89.2	61.9	48.8	57.2
3.0	95.5	63.9	58.2	59.3
3.2	102.0	64.9	56.7	61.1
3.4	108.0	72.2	62.0	62.9
3.5	111.0	75.4	70.7	64.6
3.7	118.0	82.9	67.0	66.2
3.8	121.0	81.4	69.1	67.7
4.0	127.0	Lost	70.7	68.5

TABLE IX
SUMMARY OF COMPARATIVE THERMOBALANCE CONVERSION DATA
UO₂ TO UF₄ (See Figure 11)

Time (min)	Conversion Pulverized* (%)	Conversion 60-100 Mesh (%)
0	0	0
5	67.0	40.0
10	76.0	60.0
15	80.0	75.0
20	92.0	80.0
30	94.0	89.5
40	96.0	93.0
60	97.2	95.4
80	97.9	96.8
100	99.0	97.8
140	Not Determined	99.0

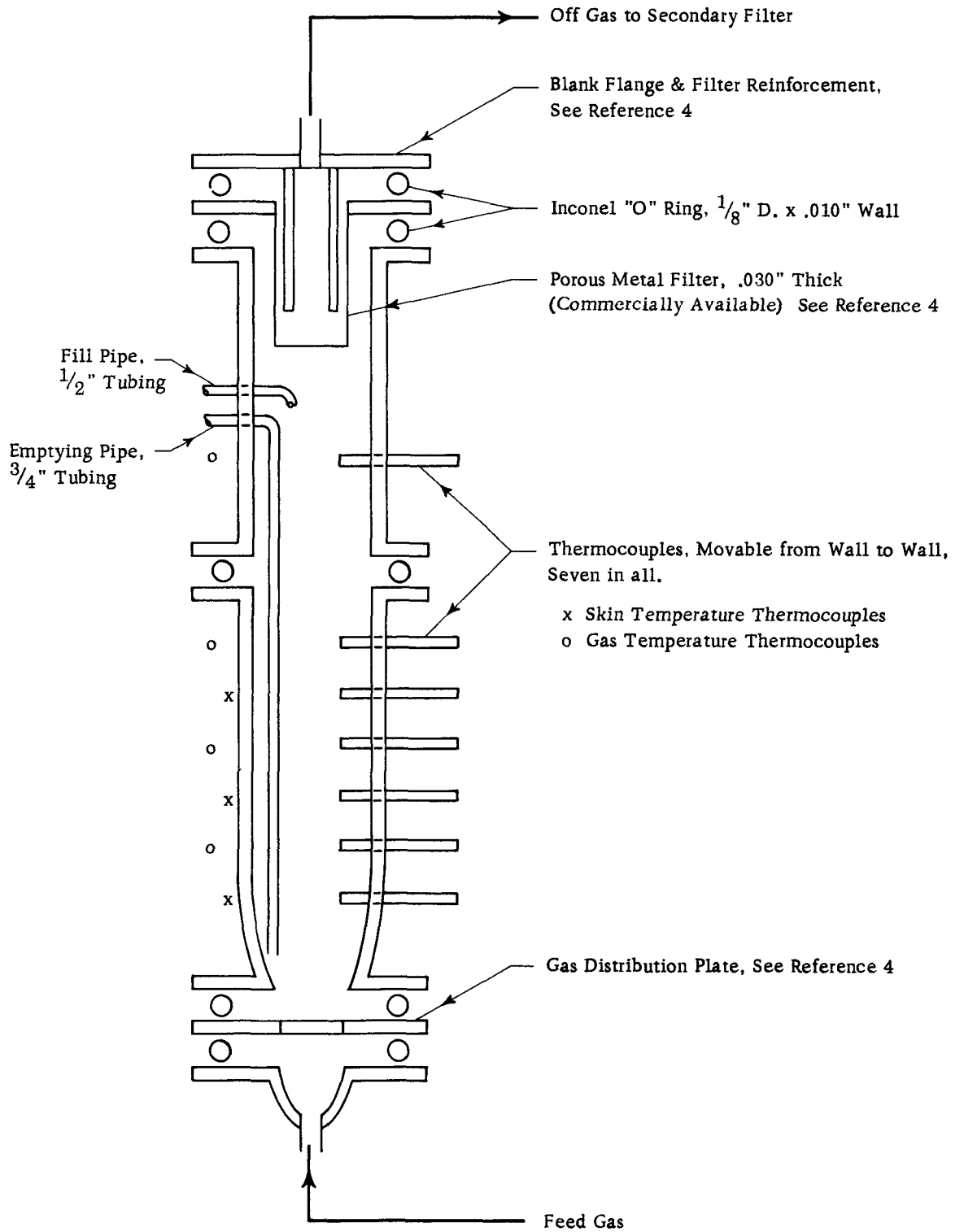
* All -200 mesh, 70% -325 mesh.

TABLE X
COMPARISON OF ESTIMATED TAPERED COLUMN PERFORMANCE WITH
EXISTING FLUIDIZED BED CONTACTORS

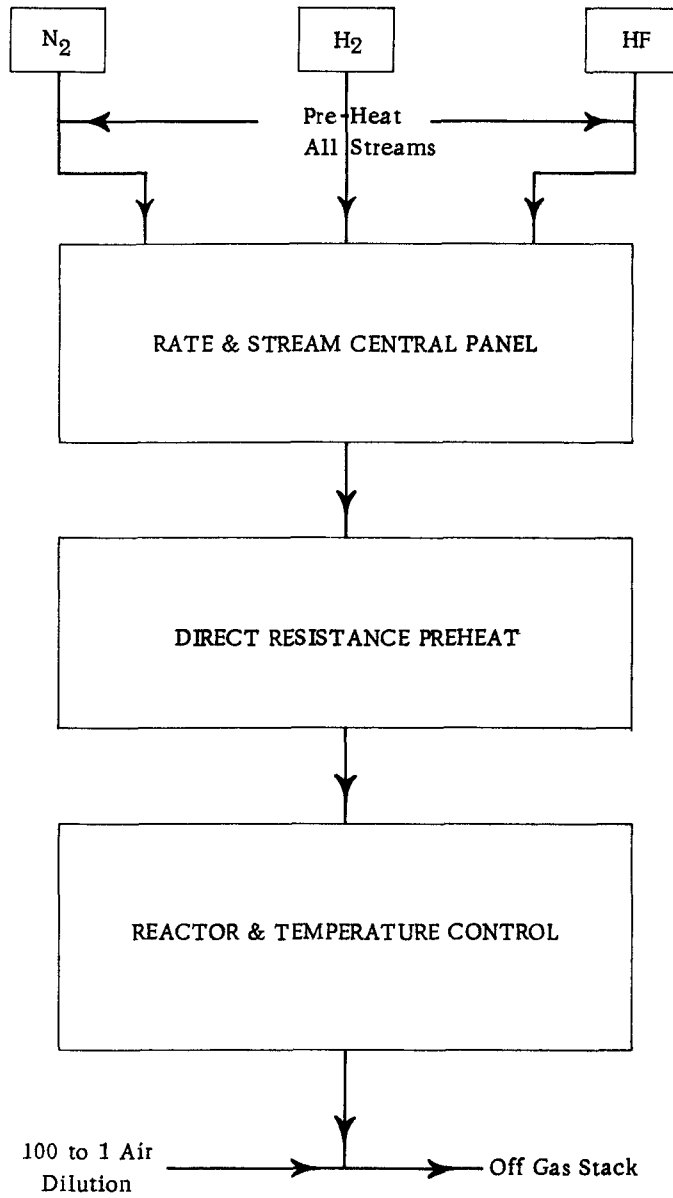
Reference Reactor	No. Stages	L/D	Productivity (lb/hr-ft ³)	Product Conversion (%)	Unit Produces	Rate Constant (hr ⁻¹)
(3)	2	3.4	60	97	UO ₂	8.0
Tapered Equivalent	1	~ 10	367	99	UO ₂	8.0
(2)	5	5.5	45	95	UF ₄	1.0
Tapered Equivalent	1	~ 10	75	95	UF ₄	1.0
(16)	5	4	80	96	UF ₄	2.4
Tapered Equivalent	1	~ 10	130	96	UF ₄	2.4

TABLE XI
SUMMARY OF DESIGN PROCEDURE FOR OPTIMIZING
A FLUIDIZED BED CONTACTOR

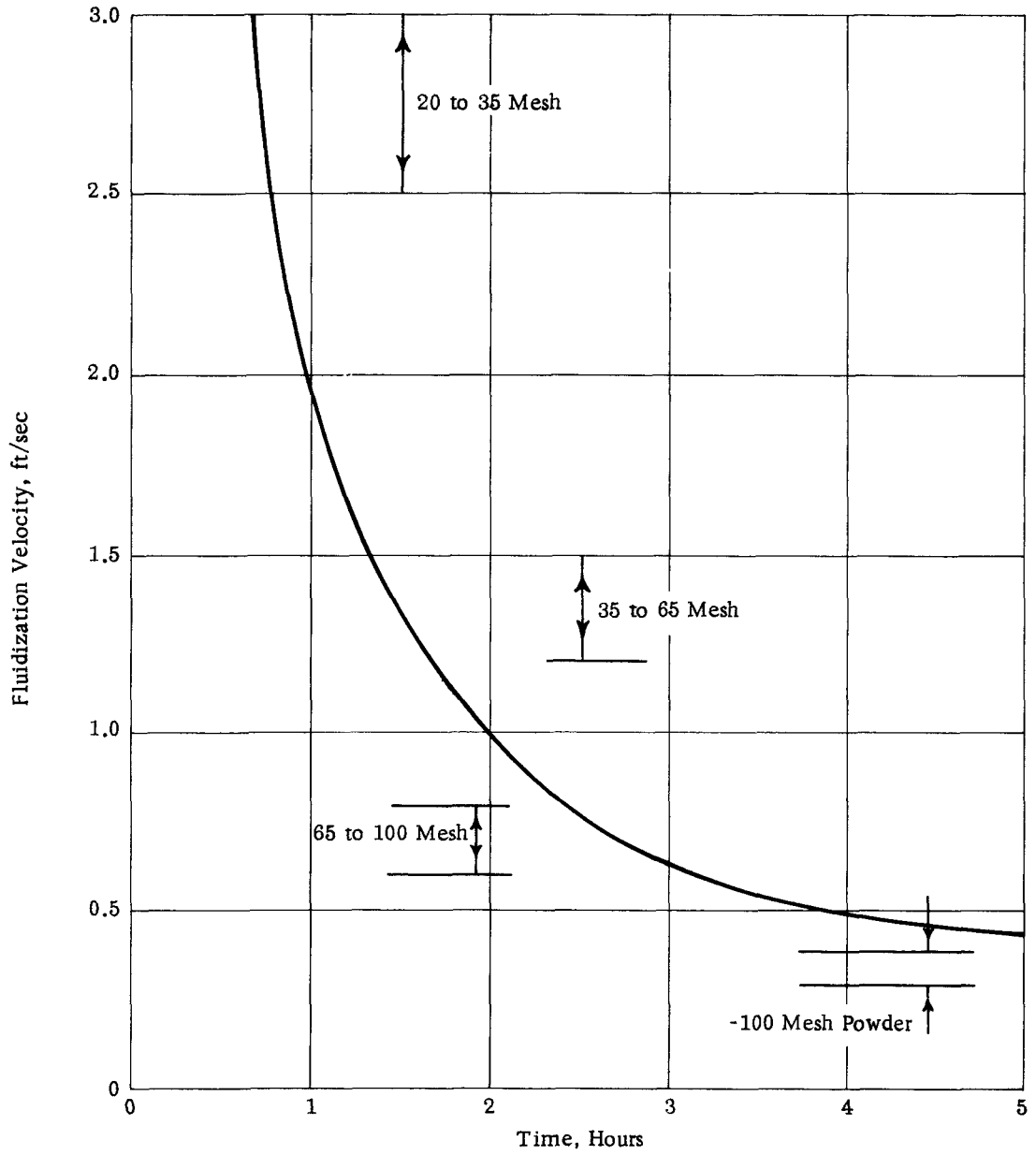
Item	Symbol	Given	Experimentally Determined	Computed
Feed Rate	F, w	x		
Conversion	c	x		
Reagent Efficiency	$P_t/P_b (100)$	x		
Particle Size or Fluidization Velocity	v	x		
Operating Void Volume	ϵ		x	
Solids Mixing Coefficient	M		x	
Reaction Rate Constant at Various Temperatures	k		x	
Required Column Taper	ϕ		x	
Heat Transfer Coefficient	h		x	
Operating Temperature Difference	Δt		x	
Optimum Temperature Gradient			x	x
Optimum Length				x
Optimum Diameter				x



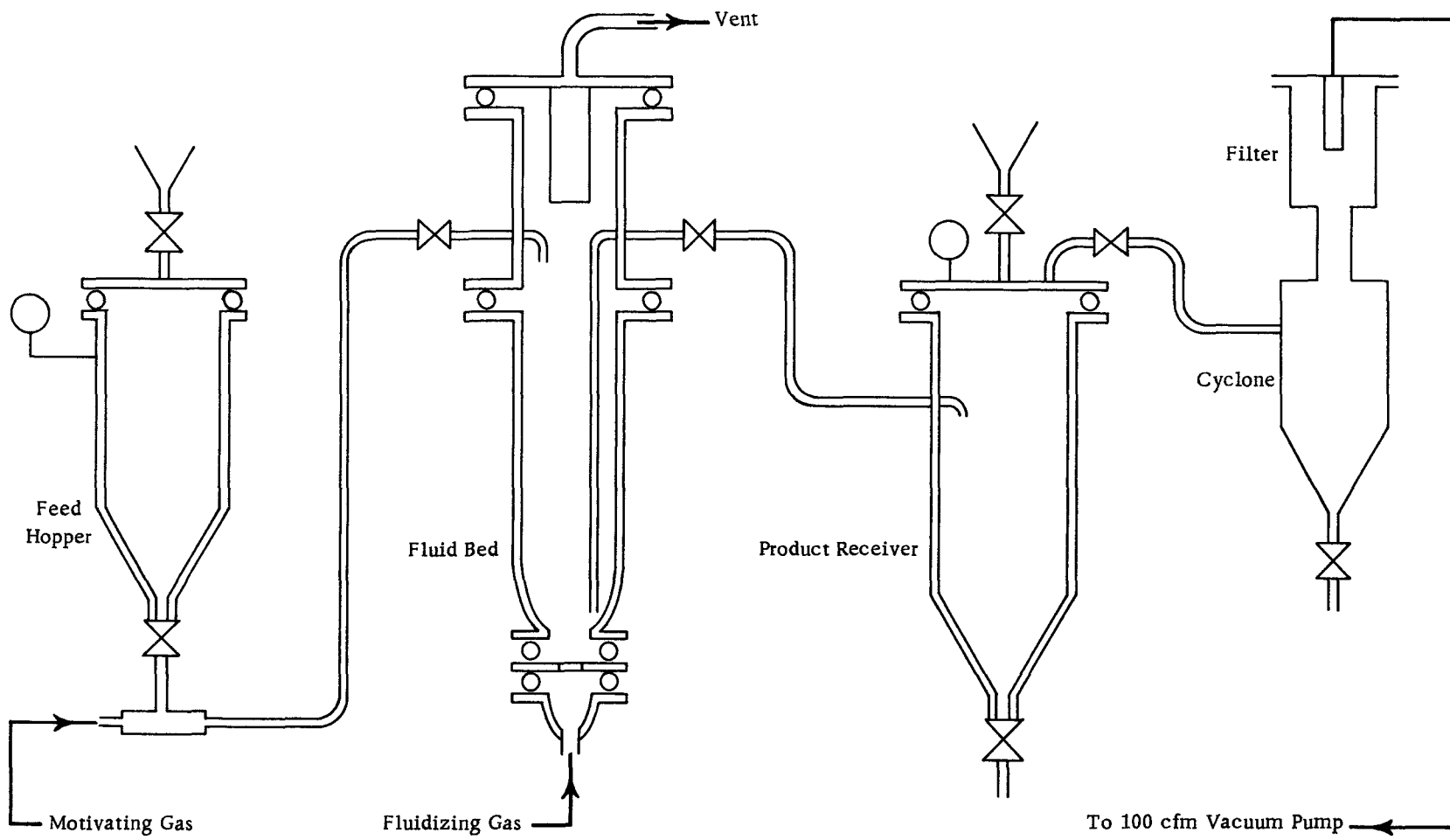
APPENDIX A - FIGURE 1. SCHEMATIC LAYOUT OF 5" FLUID BED PILOT PLANT



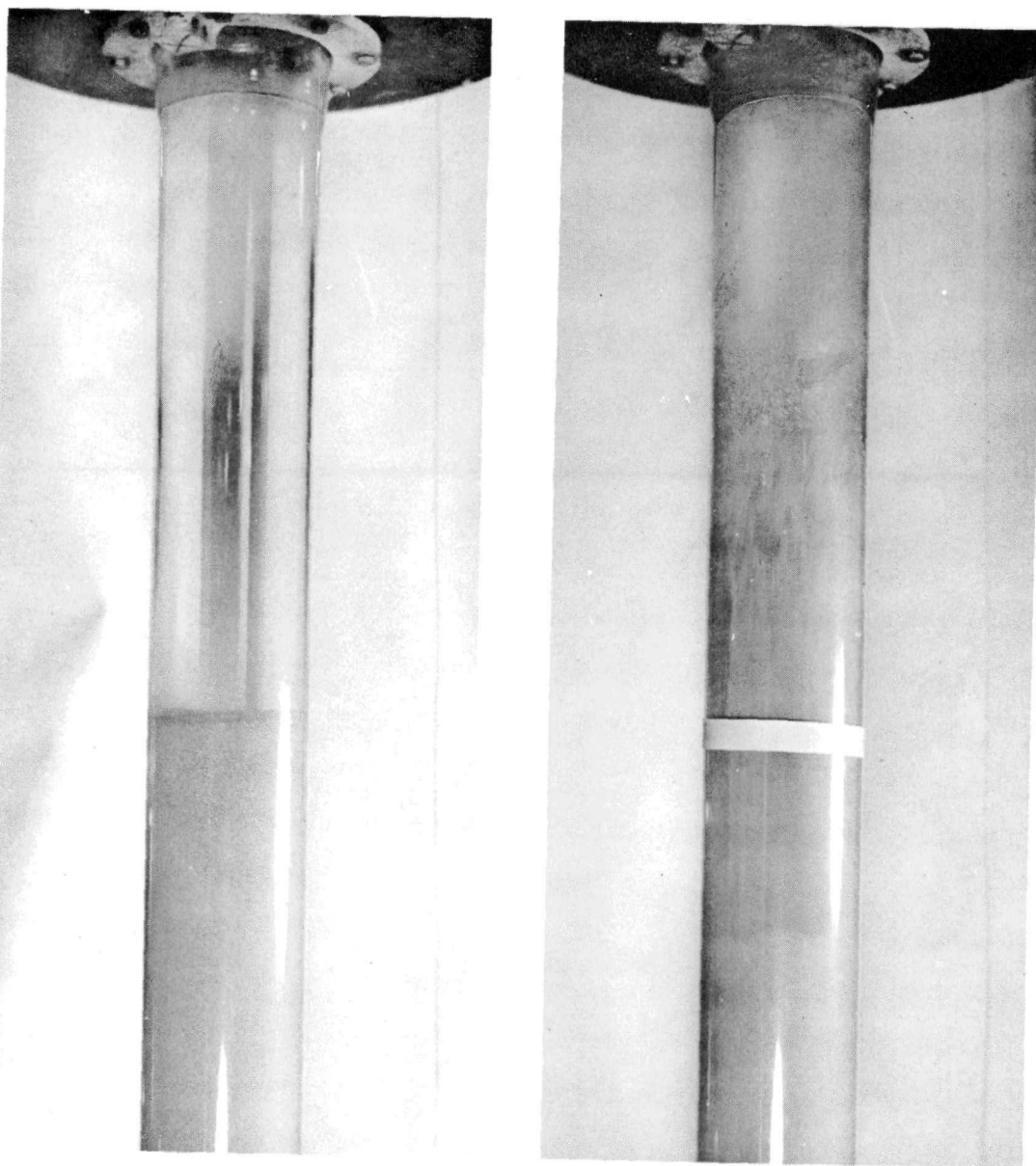
APPENDIX A - FIGURE 2. BLOCK FLOW DIAGRAM FLUID BED PILOT PLANT



APPENDIX A - FIGURE 3. FLUIDIZATION VELOCITY vs TIME REQUIRED TO PASS STOICHIOMETRIC GAS REQUIREMENT FOR 60# UO₂ IN A 5" DIA. REACTOR @1000°F



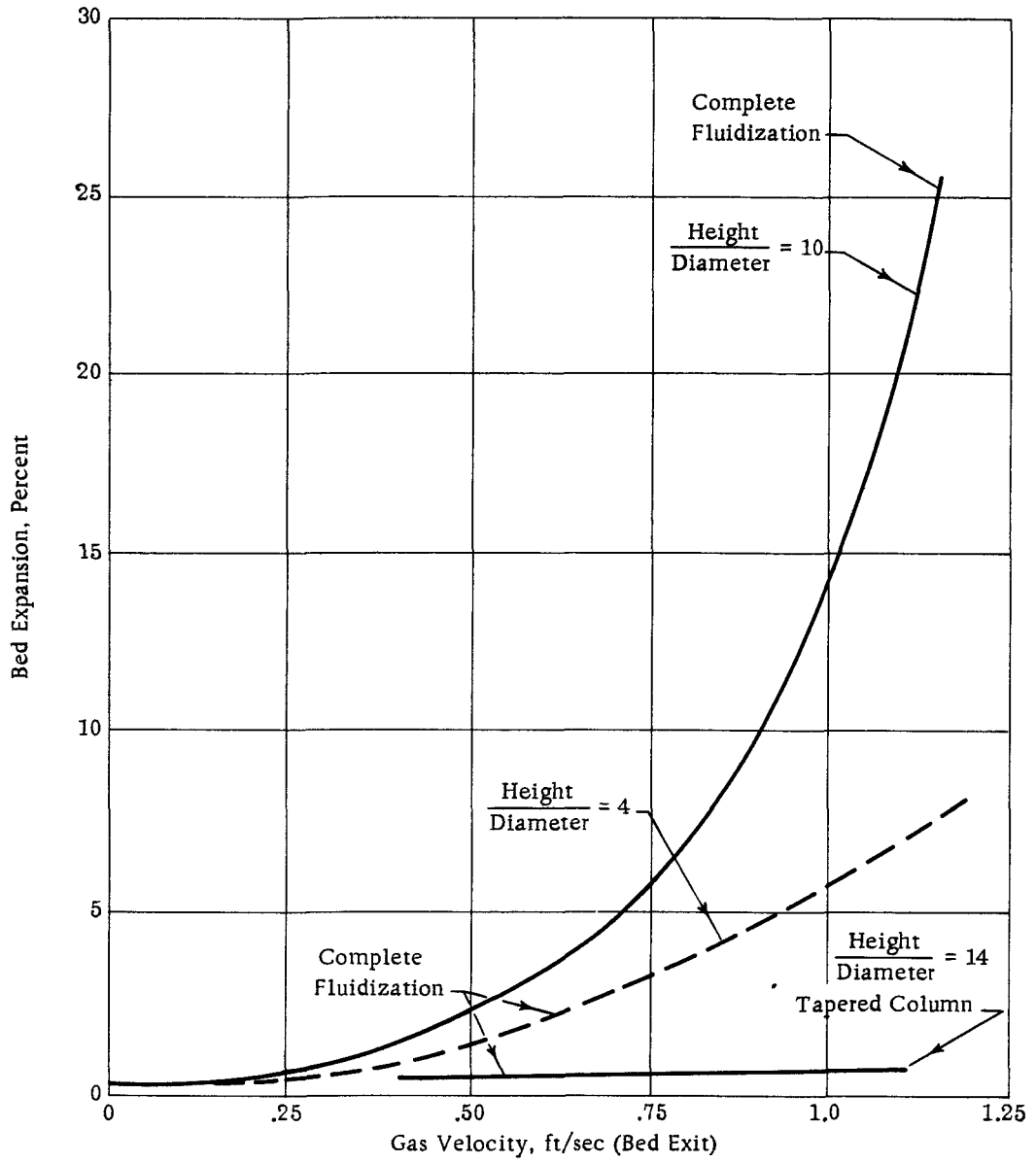
APPENDIX A - FIGURE 4. SCHEMATIC LAYOUT OF EXPERIMENTAL FILLING AND EMPTYING SYSTEM



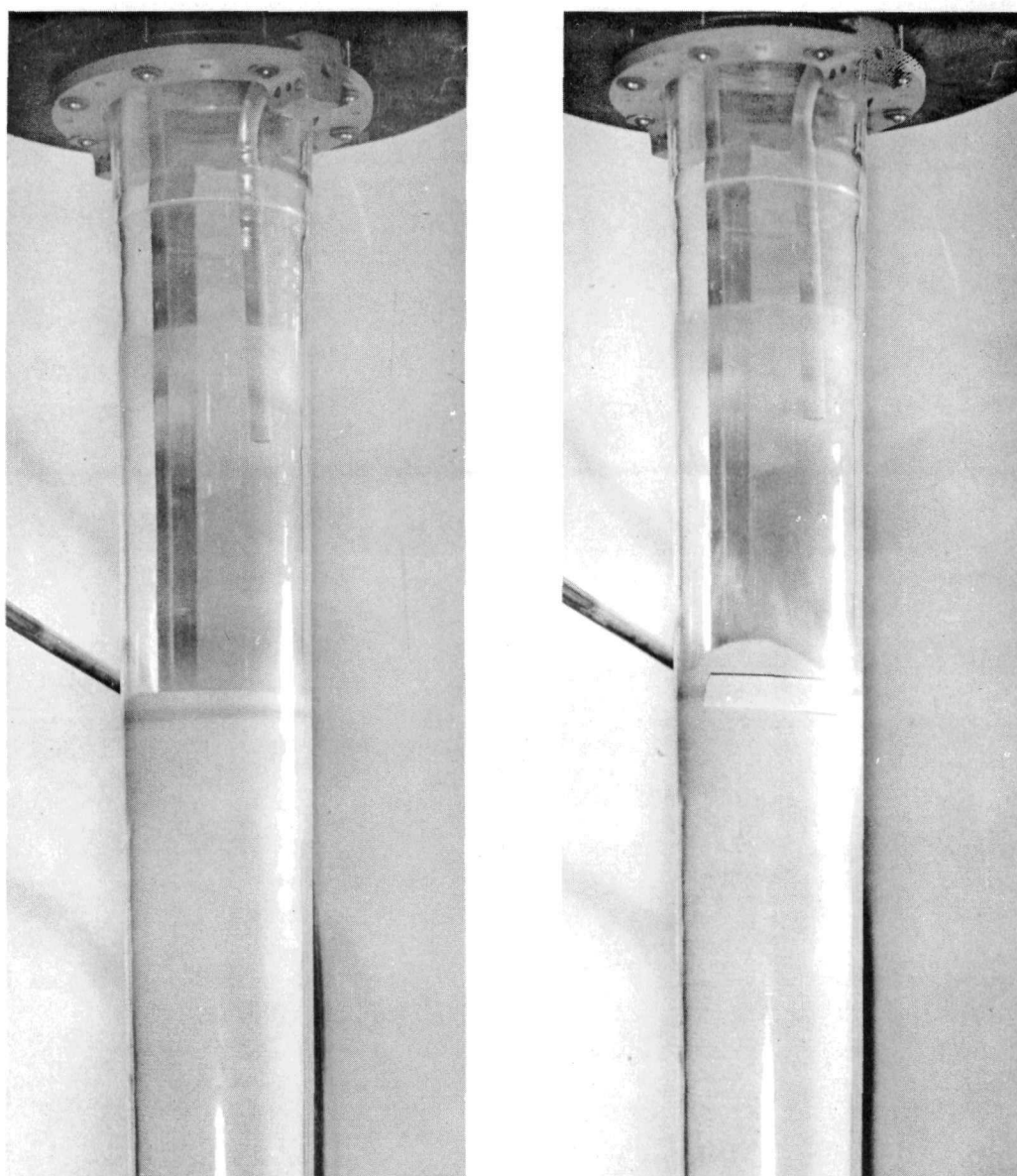
At Rest

Fluidized

FIGURE 5. PHOTOGRAPH SHOWING 20-40 MESH UO_3 POWDER
IN A 6 INCH CYLINDRICAL GLASS COLUMN
(Bed At Rest is 4 Ft. Deep)



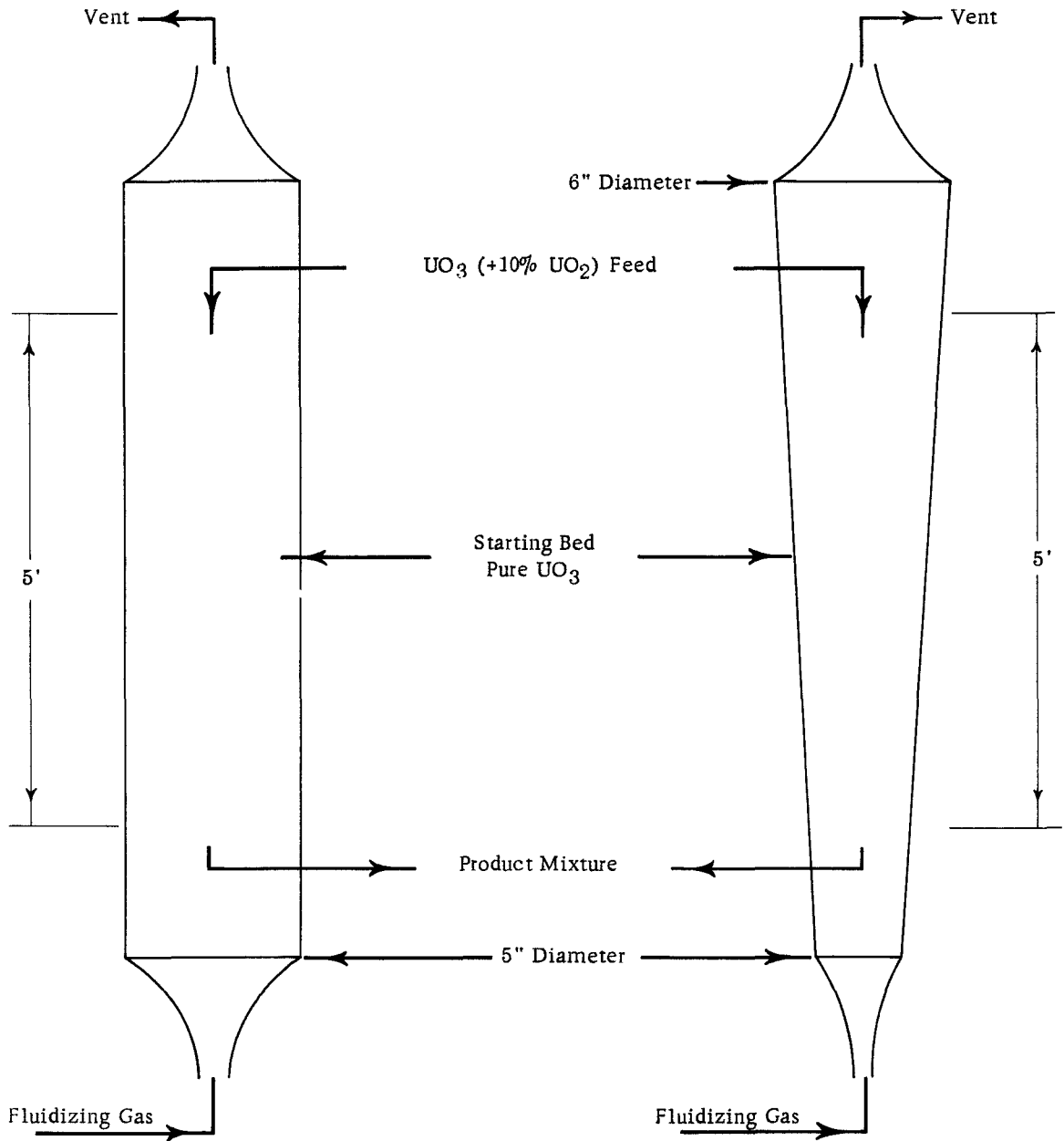
APPENDIX A - FIGURE 6. FLUIDIZATION OF -65, + 100 MESH UO_3
 BED EXPANSION vs FLUIDIZING GAS
 VELOCITY FOR TWO BED DEPTHS



At Rest

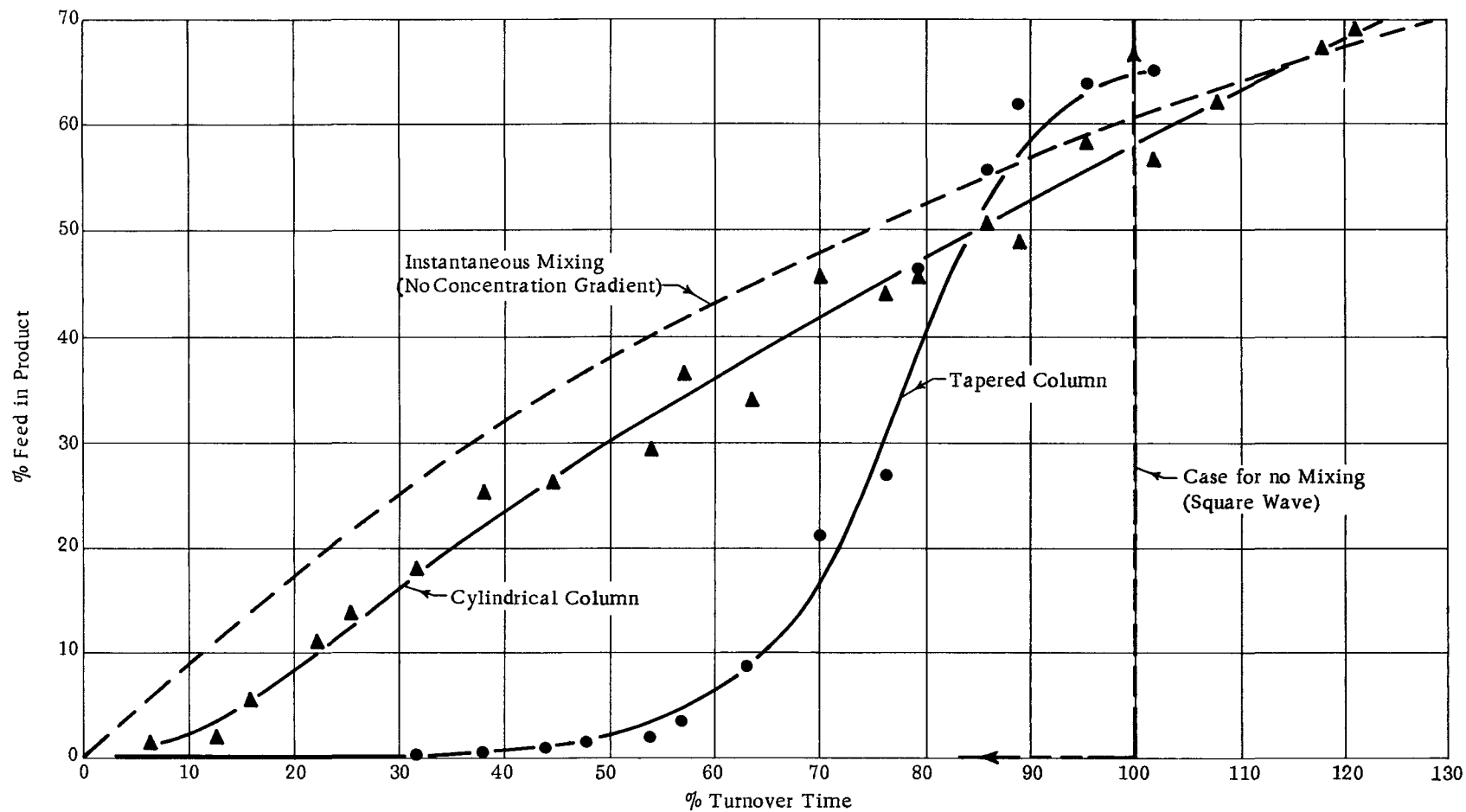
Fluidized

FIGURE 7. PHOTOGRAPH SHOWING 20-40 MESH UO_3 POWDER
IN A 6 INCH DIAMETER TAPERED COLUMN
(Bed Depth At Rest is 5 Ft.)

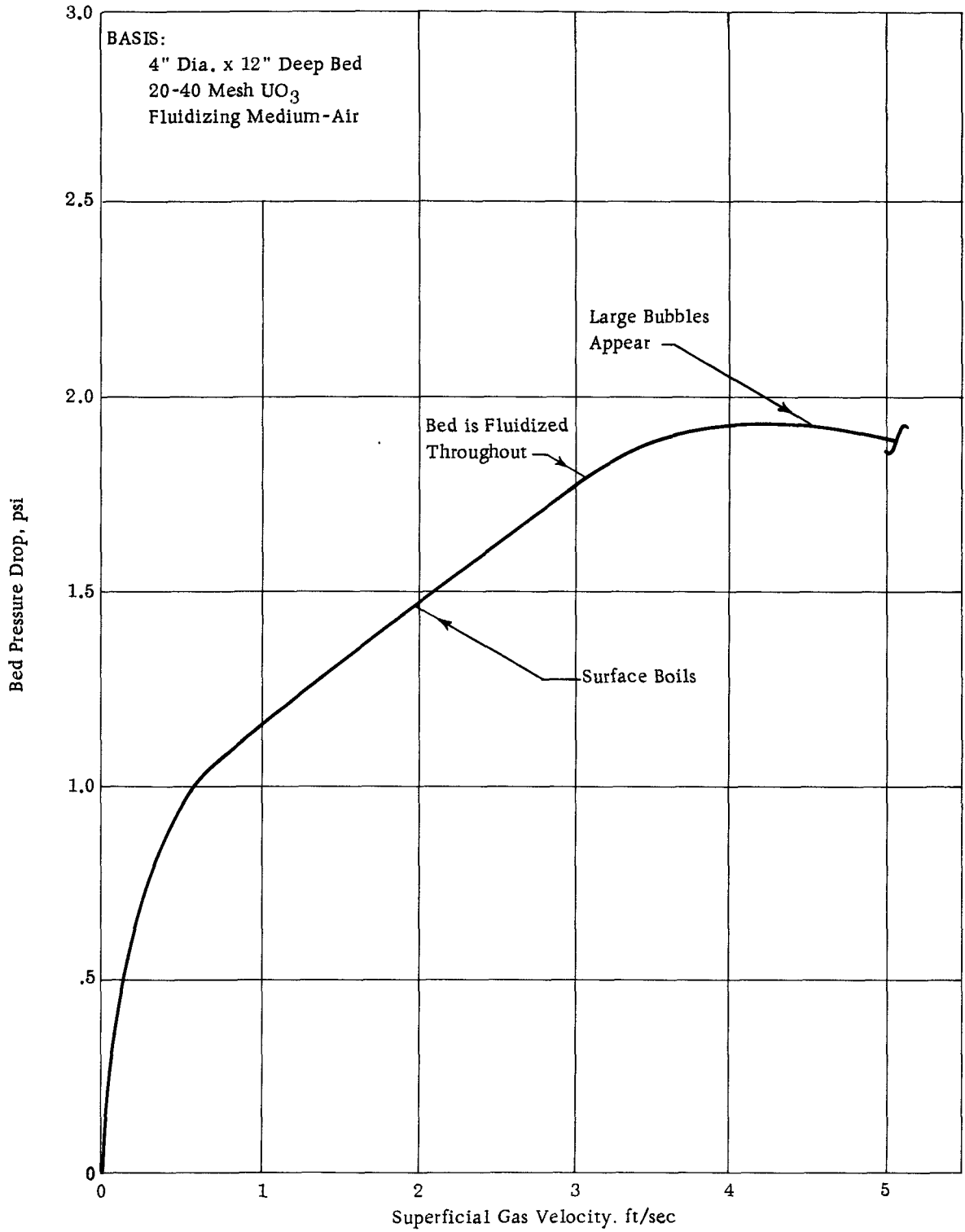


$$\text{Turnover Rate, (R)} = \frac{\text{Bed Wt. (I)}}{\text{Feed Rate (F)}} = \underline{3.14 \text{ hr.}}$$

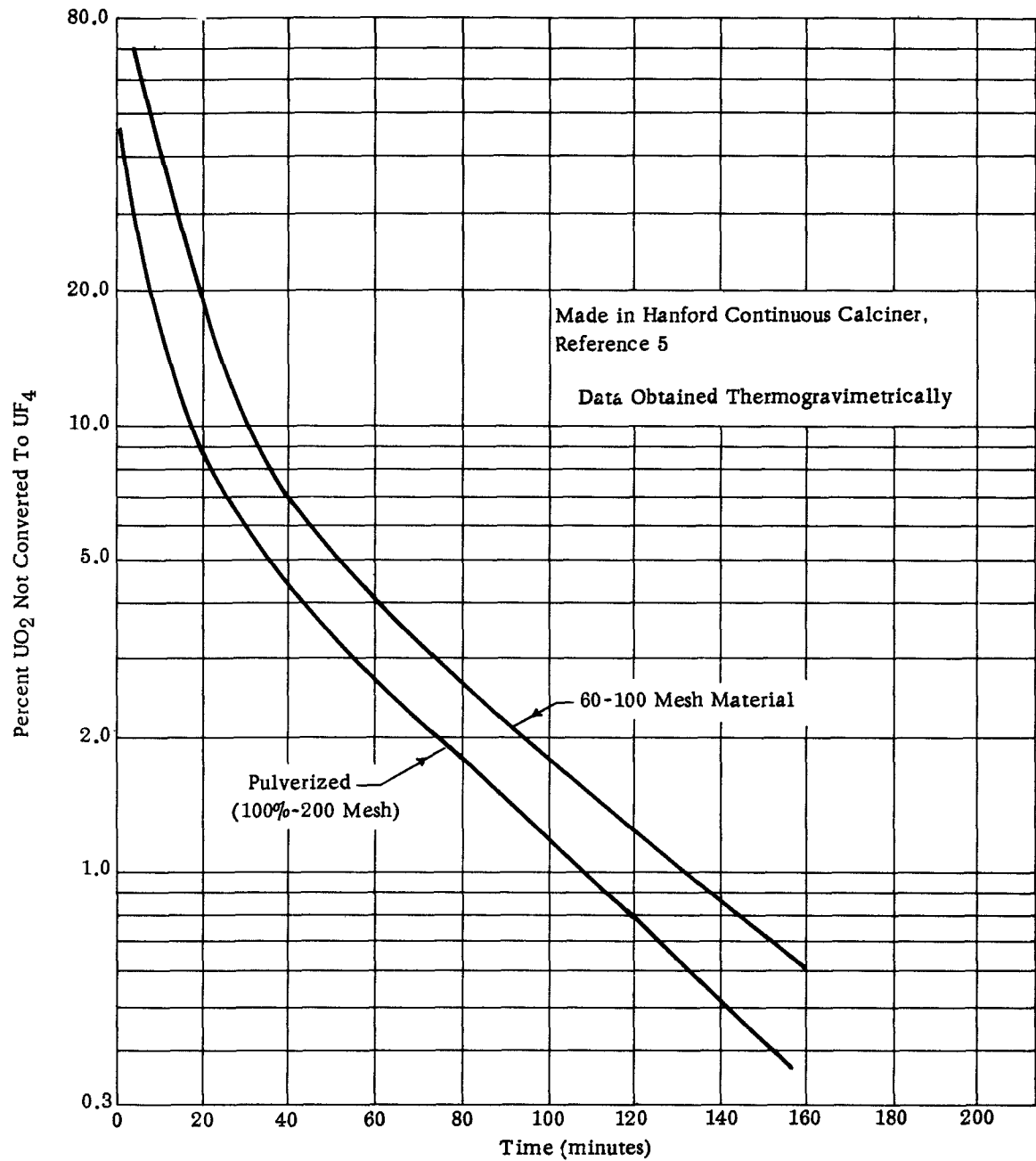
APPENDIX A - FIGURE 8. SCHEMATIC LAYOUT OF COMPARATIVE MIXING EXPERIMENT
 PRODUCT MIXTURE QUARTERED FOR DUPLICATE SAMPLES
 EVERY FIVE MINUTES



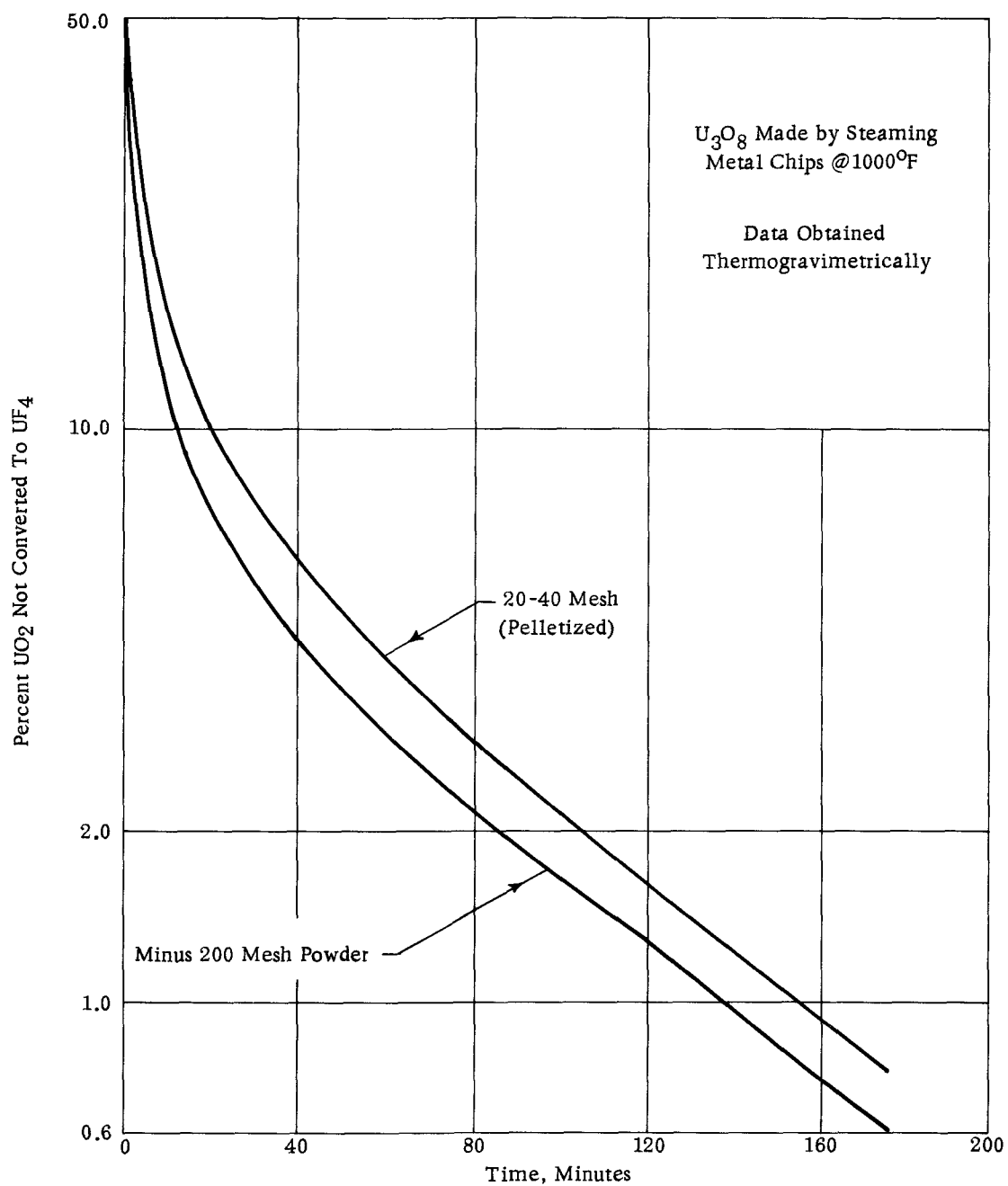
APPENDIX A - FIGURE 9. DIFFUSION OF UO_2 INTO UO_3 IN A CONTINUOUS FLUID BED OPERATION
 CONCENTRATION OF FEED IN PRODUCT vs % TURNOVER TIME
 IN BOTH A CYLINDRICAL AND A TAPERED BED
 (Turnover Time = 3.14 Hr.)



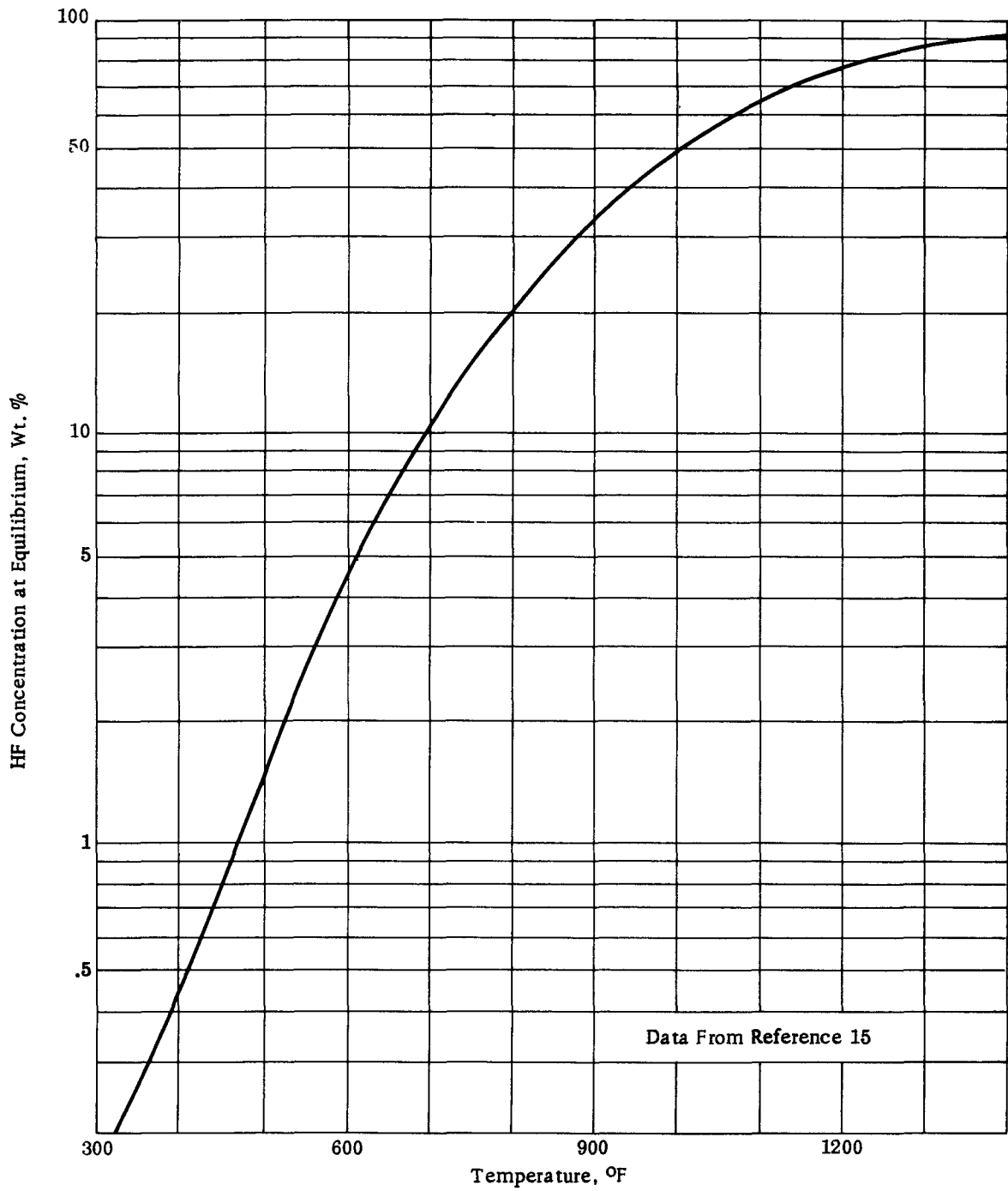
APPENDIX A - FIGURE 10. BED PRESSURE DROP vs GAS VELOCITY



APPENDIX A - FIGURE 11. % UNCONVERTED UO_2 vs TIME
 UO_3 FEED REDUCED @1040°F
HYDROFLUORINATED @932°F



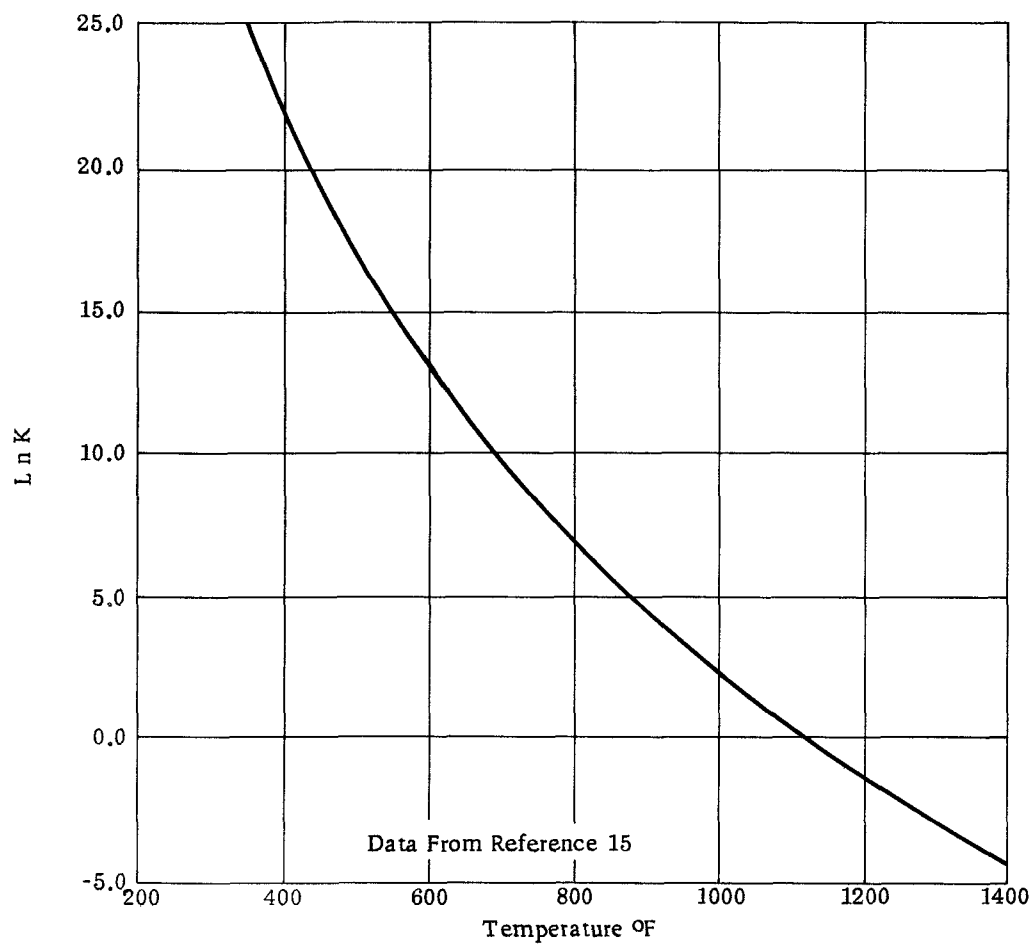
APPENDIX A - FIGURE 11A. % UNCONVERTED UO_2 vs TIME
 U_3O_8 FEED REDUCED @1040°F
HYDROFLUORINATED @1022°F



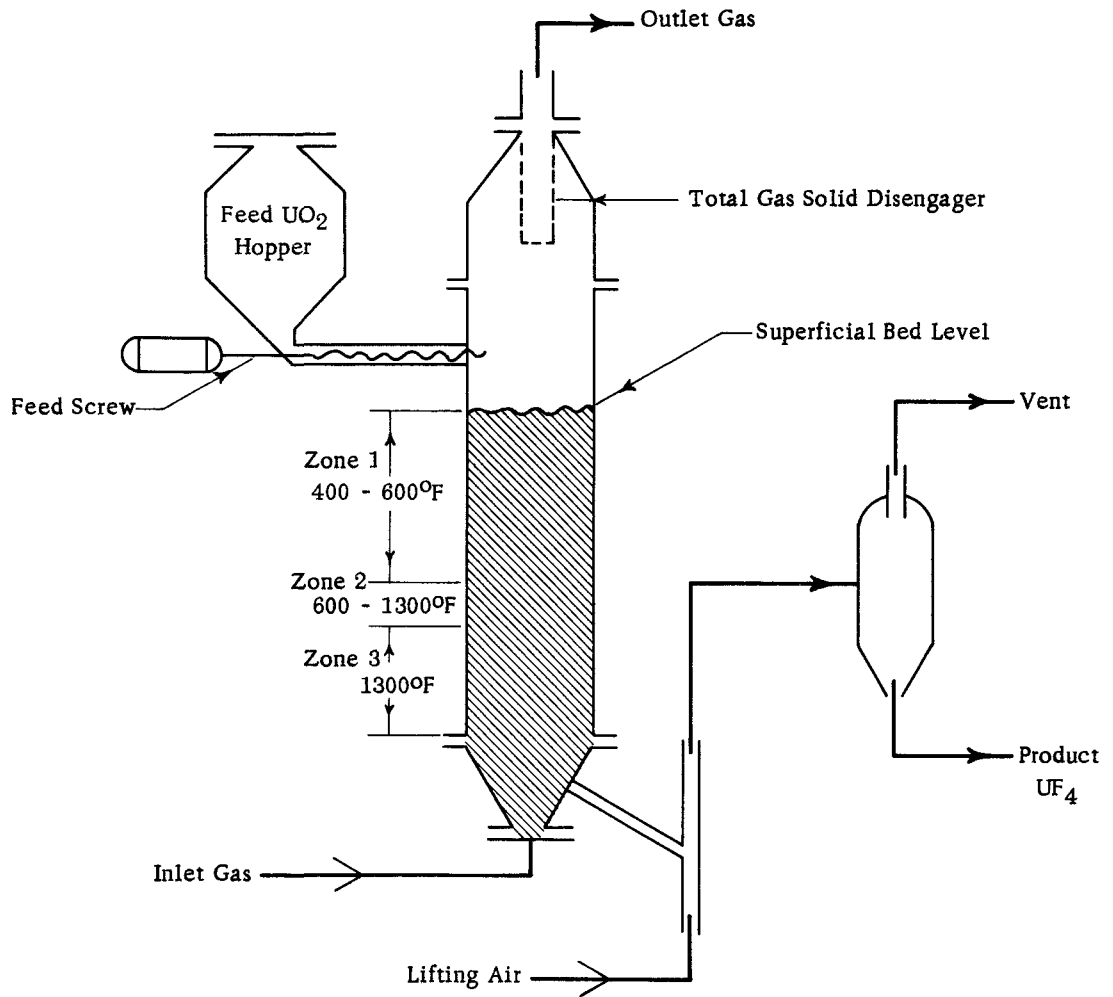
APPENDIX A - FIGURE 12. CONCENTRATION OF HF (Wt. %) IN EQUILIBRIUM WITH A MIXTURE

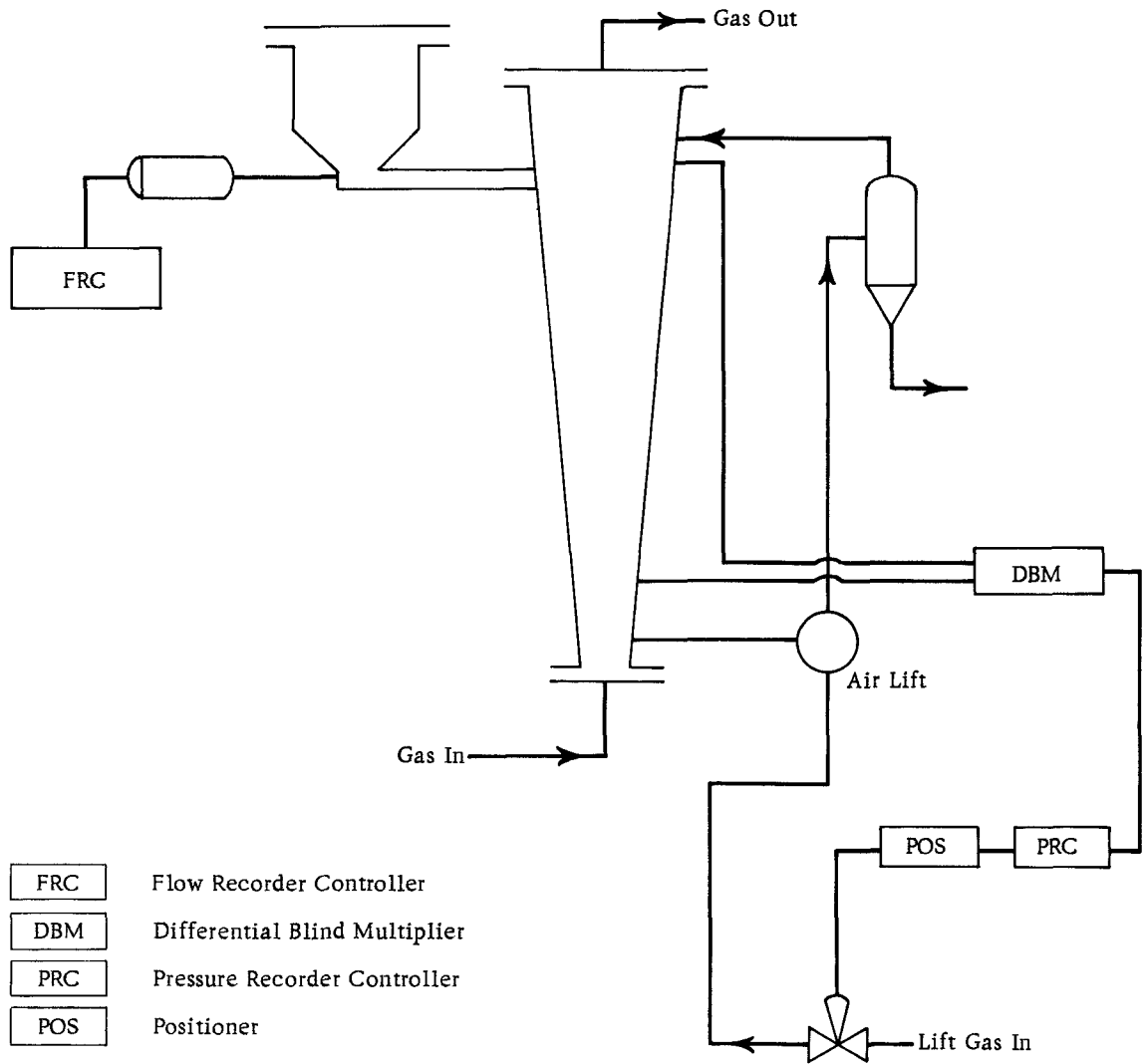
UO₂ & UF₄

$$P_T = P_{HF} + P_{H_2O}$$

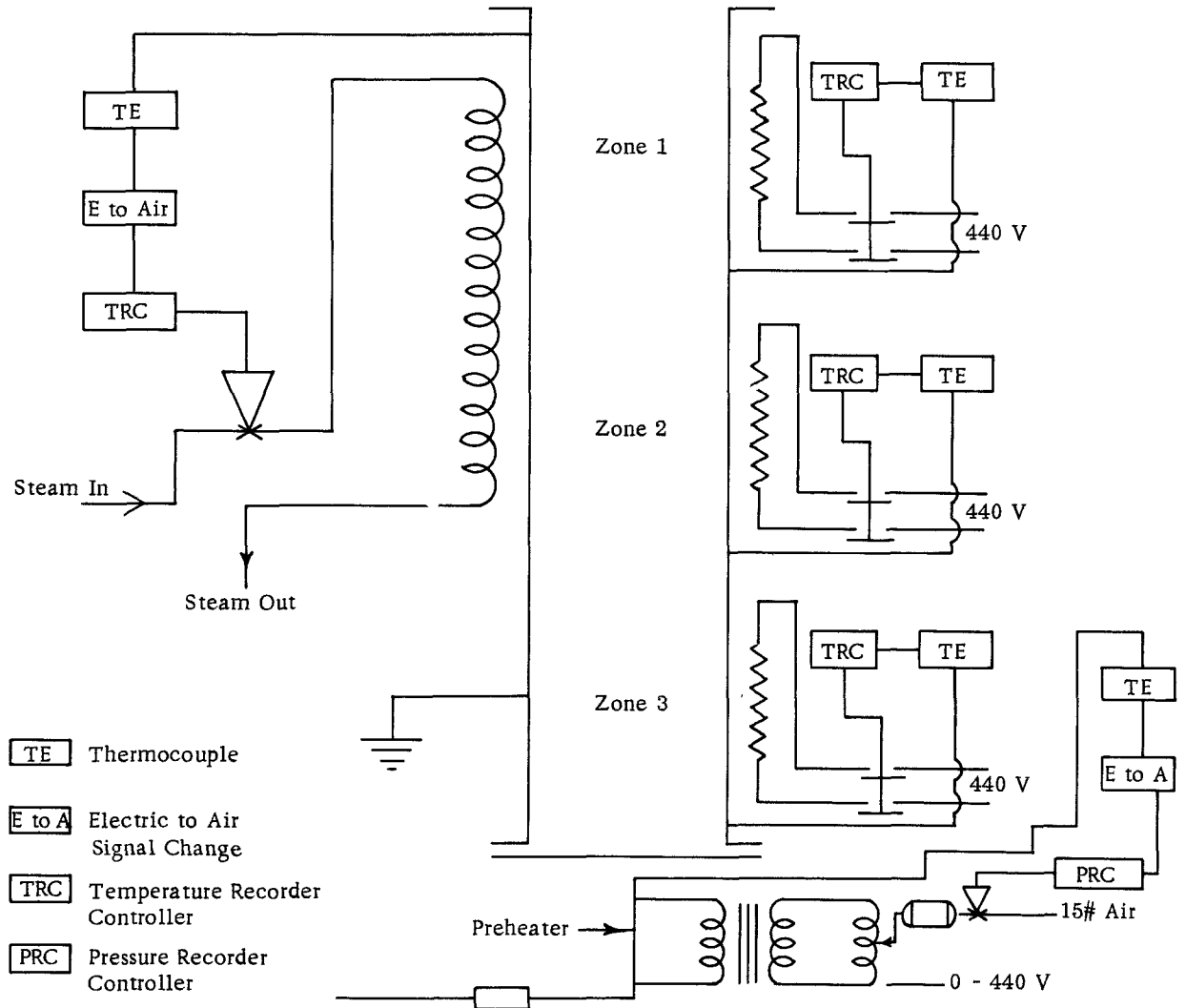


APPENDIX A - FIGURE 13. NATURAL LOG K (FOR $\text{UO}_2 + 4\text{HF} \rightleftharpoons \text{UF}_4 + 2\text{H}_2\text{O}$)
vs TEMPERATURE

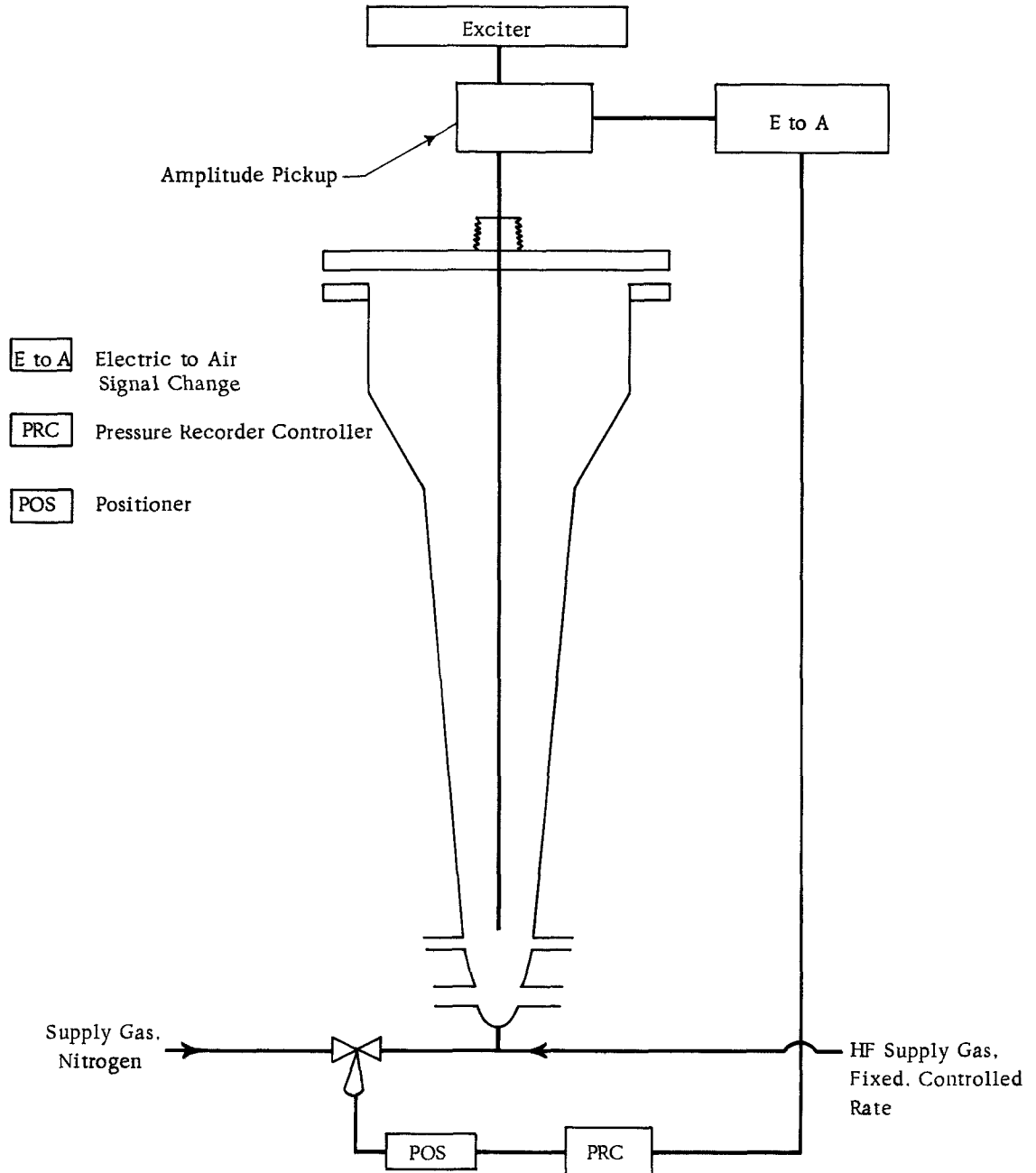
APPENDIX A - FIGURE 14. TYPICAL COLUMN DESIGN FOR UF_4 PRODUCTION



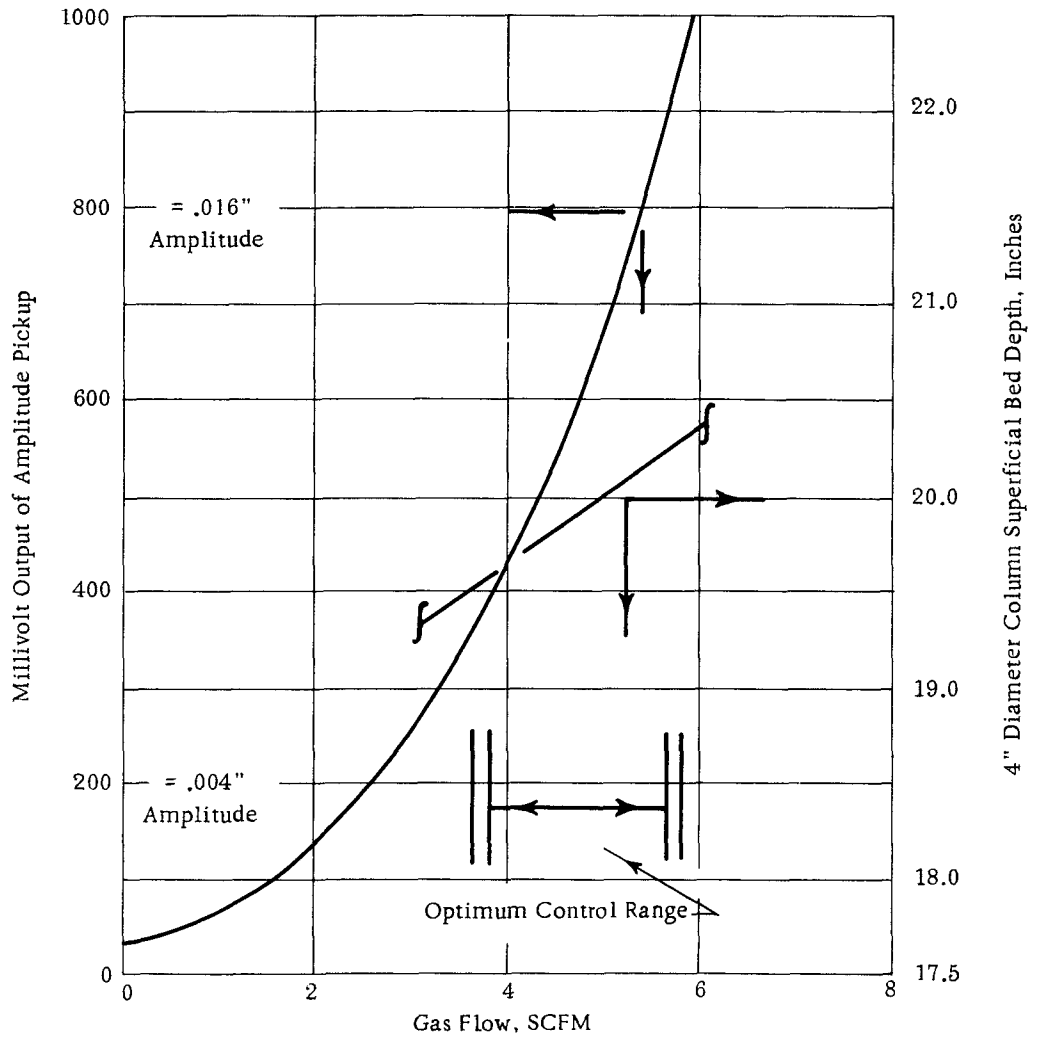
APPENDIX A - FIGURE 15. SCHEMATIC INSTRUMENT LAYOUT FOR TYPICAL TAPERED FLUID BED INVENTORY CONTROL



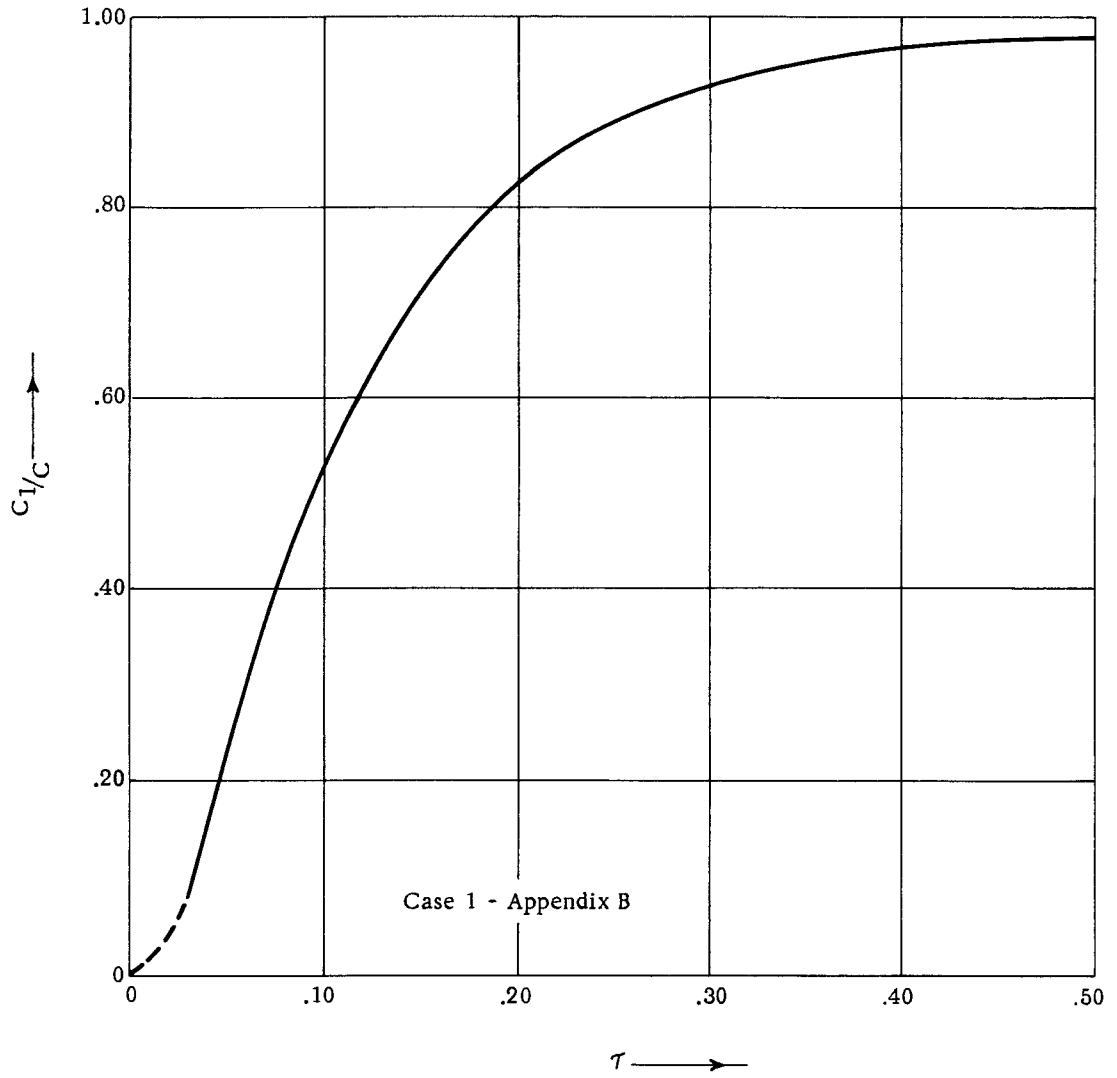
APPENDIX A - FIGURE 16. SCHEMATIC LAYOUT FOR TYPICAL TAPERED FLUID BED HEATING AND COOLING CONTROL



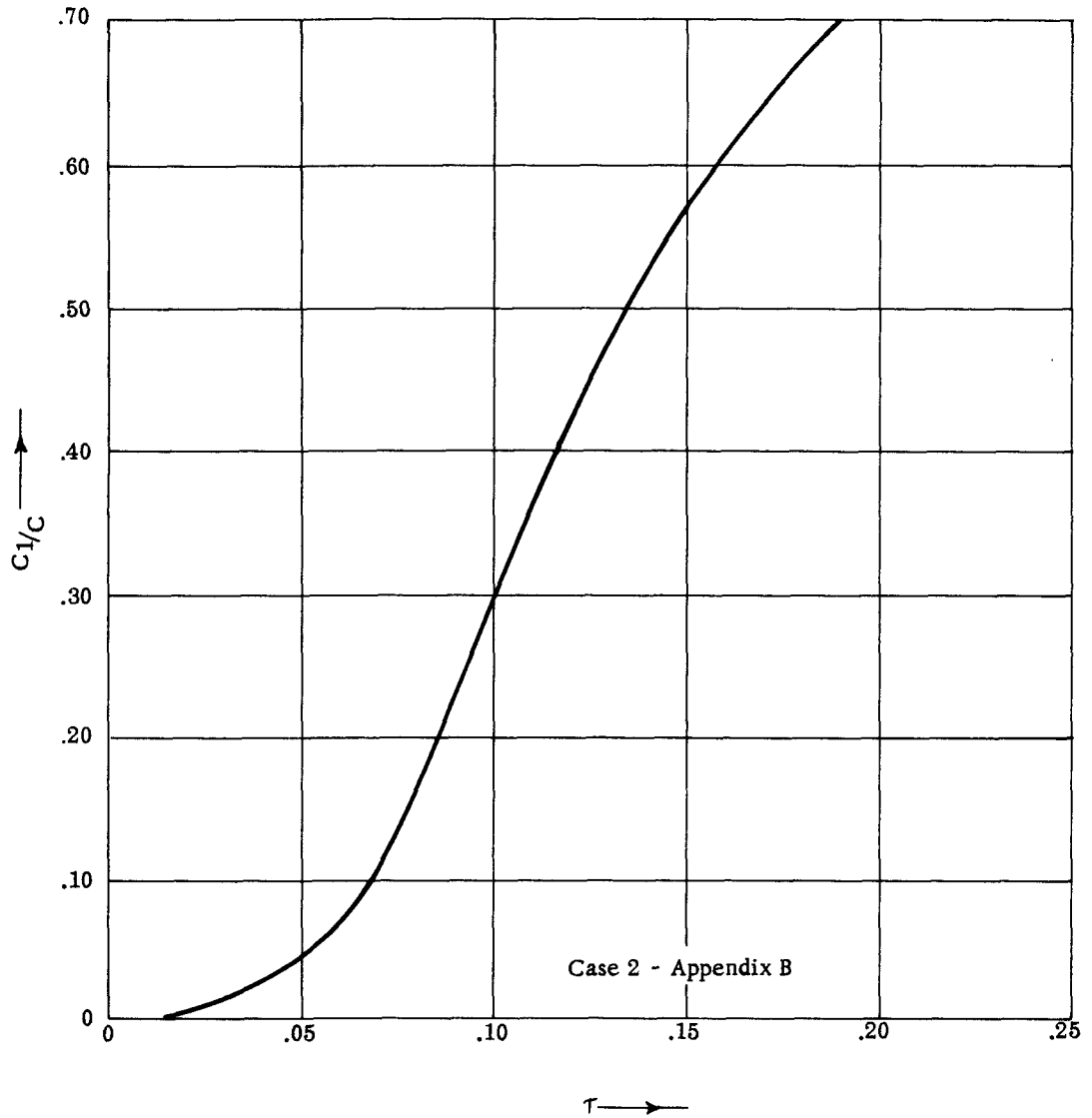
APPENDIX A - FIGURE 17. SCHEMATIC LAYOUT FOR TYPICAL TAPERED COLUMN FLUIDIZATION CONTROLLER



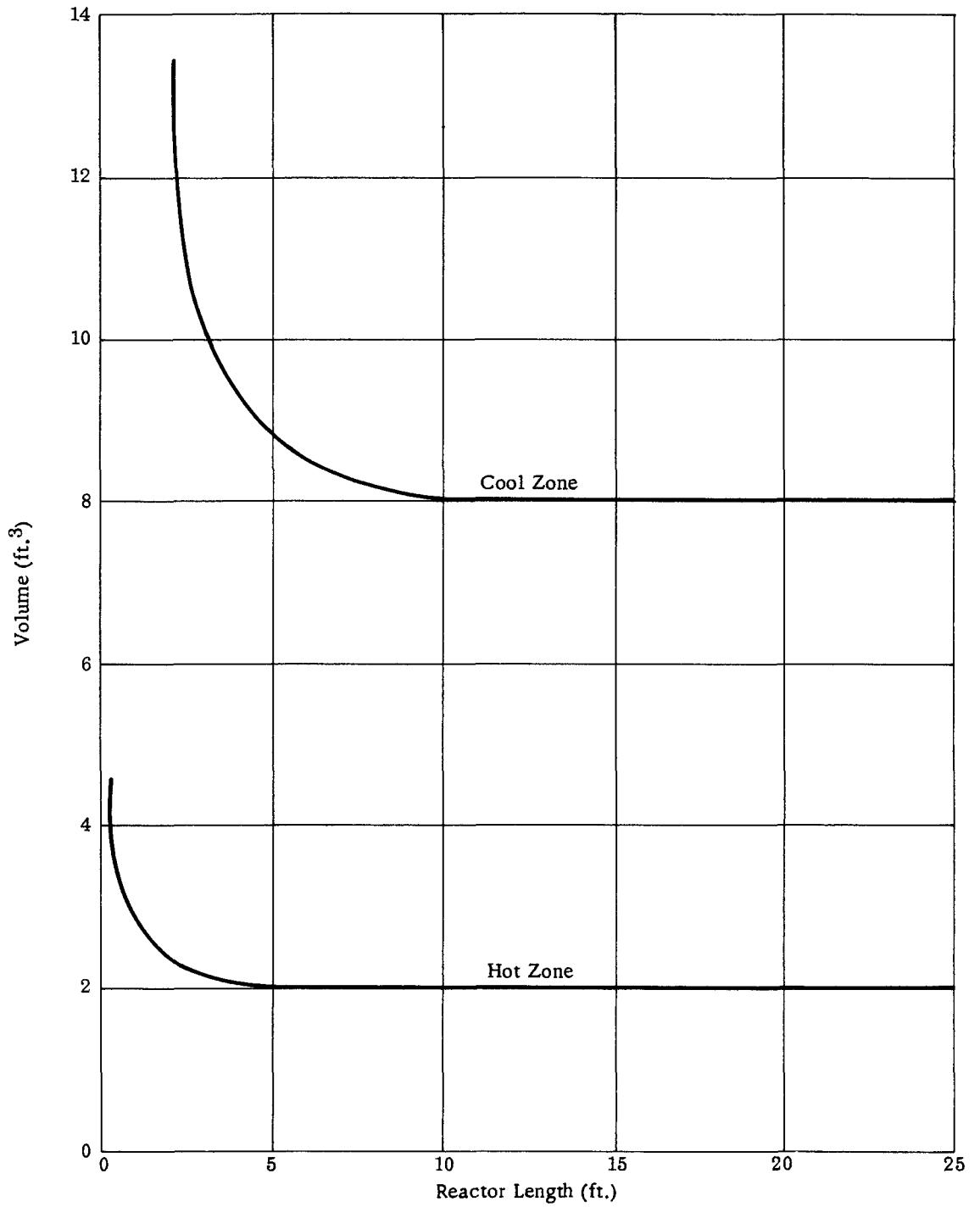
APPENDIX A - FIGURE 18. FLUIDIZATION CONTROLLER OUTPUT vs GAS FLOW



APPENDIX A - FIGURE 19. PLOT FOR DETERMINING THE MIXING COEFFICIENT "M" WHEN STARTING A FLUIDIZED BED OF TWO EQUAL LAYERS OF MATERIALS OF EQUAL DENSITY AND PARTICLE SIZE



APPENDIX A - FIGURE 20. PLOT FOR DETERMINING MIXING COEFFICIENT "M" (See Appendix B)
WHEN A TRACE OF DIFFUSING MATERIAL IS INJECTED IN A
FLUIDIZED BED OF ANOTHER MATERIAL OF THE SAME
DENSITY AND PARTICLE SIZE

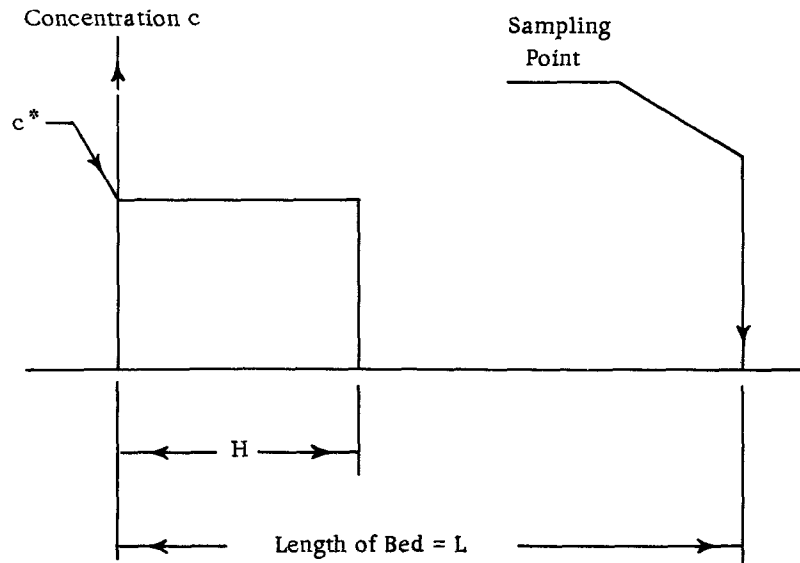


APPENDIX A - FIGURE 21. COLUMN VOLUME vs COLUMN LENGTH FOR THE COLUMN DESIGN OF APPENDIX E SECTION 3

APPENDIX BDETERMINATION OF AN EXPERIMENTAL MIXING COEFFICIENT

The derivation of a mixing coefficient in which constant column mixing is assumed will be presented.

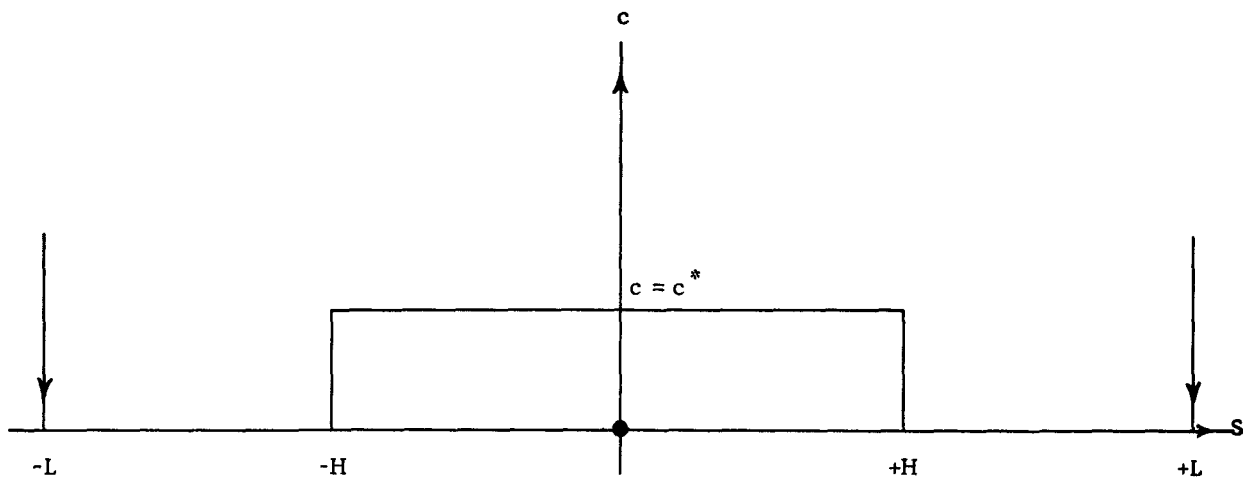
Consider a uniform bed of powder (the taper in the tapered columns is so slight that it is ignored) of constant density and initial inventory as follows:



As time goes by, samples are taken at the sampling point at one extreme of the bed, and from the curve of the sample concentrations versus time, the mixing coefficient is determined.

MATHEMATICAL SOLUTION

For convenience, it is better to consider the setup of the above sketch as half of the symmetrical setup shown below:



The differential equation which must be satisfied is

$$M \frac{\partial^2 c}{\partial s^2} = \frac{\partial c}{\partial t}, \quad (1)$$

where

$c = c(s, t)$ is the concentration of one of the powder components,

$s =$ distance,

$t =$ time,

$M =$ mixing coefficient.

The boundary conditions which must also be satisfied are:

$$\frac{\partial c}{\partial s} = 0 \quad \left\{ \begin{array}{l} \text{for } s = -L, t > 0, \\ \text{for } s = L, t > 0, \end{array} \right. \quad (2)$$

(3)

and

$$\left. \begin{array}{l} c = c^* \quad \text{for } -H \leq s \leq H, t = 0, \\ c = 0 \quad \text{for } -L < s < -H, \\ \quad \quad \quad H < s \leq L, t = 0. \end{array} \right\} \quad (4)$$

Conditions (2) and (3) result from demanding no material transfer at the extremes $s = -L$ and $s = L$, and condition (4) describes the initial loading of the bed.

For convenience, we let $\sigma = s/L$ and $\tau = Mt/L^2$, and with this change of variables, (1) through (4) are written:

$$\frac{\partial^2 c}{\partial \sigma^2} = \frac{\partial c}{\partial \tau}, \quad (5)$$

$$\frac{\partial c}{\partial \sigma} = 0 \quad \left\{ \begin{array}{l} \text{for } \sigma = -1, \tau > 0, \\ \text{for } \sigma = 1, \tau > 0, \end{array} \right. \quad (6)$$

(7)

$$\left. \begin{array}{l} c = c^* \quad \text{for } -H/L \leq \sigma \leq H/L, \quad \tau = 0, \\ c = 0 \quad \text{for } -1 \leq \sigma \leq -H/L, \\ \quad \quad \quad H/L < \sigma < 1, \quad \tau = 0. \end{array} \right\} \quad (8)$$

The solution to (5) is

$$c(\sigma, \tau) = C_0 + \sum_{n=1}^{\infty} (A_n \sin \lambda_n \sigma + B_n \cos \lambda_n \sigma) e^{-\lambda_n^2 \tau} \quad (9)$$

where C_0 , A_n , B_n , and λ_n are constants. To satisfy (6) and (7), we have that $A_n = 0$ and $\lambda_n = n\pi$ so that

$$c = C_0 + \sum_{n=1}^{\infty} B_n \cos n\pi\sigma e^{-(n\pi)^2 \tau} \quad (10)$$

At $\tau = 0$, we have

$$c_0 = C_0 + \sum_{n=1}^{\infty} B_n \cos n\pi\sigma \quad (11)$$

and C_0 and B_n must be chosen to satisfy (8).

We have from (8) and (11)

$$\int_{-1}^1 c_0 d\sigma = 2c^*H/L = \int_{-1}^1 C_0 d\sigma = 2C_0,$$

so that $C_0 = c^*H/L$, and for $m \neq n$,

$$\int_{-1}^1 c_0 \cos m\pi\sigma = \frac{2c^* \sin m\pi H/L}{m\pi} = B_m, \quad m \geq 1,$$

so that $B_m = 2(c^*H/L) \frac{\sin m\pi H/L}{m\pi H/L}$. Hence, we have from these results and from (10) that

$$c = (c^*H/L) \left[1 + 2 \sum_{n=1}^{\infty} \frac{\sin n\pi H/L}{n\pi H/L} \cos n\pi\sigma e^{-(n\pi)^2 \tau} \right]. \quad (12)$$

We are particularly interested in the behavior of c at the sampling point, that is, $c = c_1$ at $\sigma = 1$ we have that

$$c_1 = (c^*H/L) \left[1 + 2 \sum_{n=1}^{\infty} \frac{\sin n\pi H/L}{n\pi H/L} (-1)^n e^{-(n\pi)^2 \tau} \right]. \quad (13)$$

It is clear that $c^*_{H/L}$ must be the average concentration \bar{c} , so that finally, we have

$$c_1/\bar{c} = 1 + 2 \sum_{n=1}^{\infty} \frac{\sin n\pi H/L}{n\pi H/L} (-1)^n e^{-(n\pi)^2 \tau} \quad (14)$$

where

$$\tau = Mt/L^2 .$$

EVALUATION FOR TWO CASES OF INTEREST

Case 1:

In this case one half of the bed is initially loaded up with one component, and the other half with the other component, so that as shown in Figure 19, $H/L = 1/2$. Equation (14) then becomes

$$c_1/\bar{c} = 1 + \frac{4}{\pi} (-e^{-\pi^2 \tau} + 1/3 e^{-(3\pi)^2 \tau} - 1/5 e^{-(5\pi)^2 \tau} + 1/7 e^{-(7\pi)^2 \tau} - \dots)$$

c_1/\bar{c} versus τ is plotted in Figure 19 for this case.

Case 2:

Another case of interest is when the bed is initially loaded up with only one component, and at time $t = 0$, a small amount of the other component is introduced at one end of the bed, opposite from the sampling point. In Figure 20 it is seen that this case is approximated by letting $H/L \rightarrow 0$. In (14), we then have that $(\sin n\pi H/L) / n\pi H/L \rightarrow 1$, so that

$$c_1/\bar{c} = 1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-(n\pi)^2 \tau} . \quad (15)$$

c_1/\bar{c} versus τ is plotted in Figure 20 for this case.

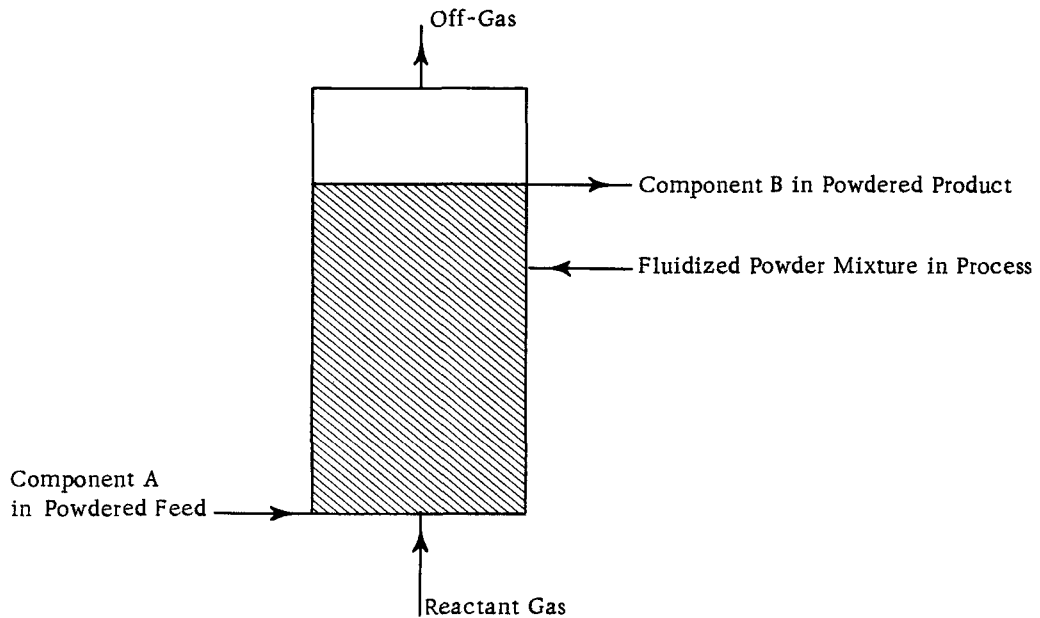
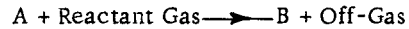
APPENDIX C

MIXING CONSIDERATIONS IN A FLUIDIZED BED

INTRODUCTION

The productivity of a fluidized bed column with constant mixing and a first order reaction in progress is considered.

The fluidized bed system, depicted in the sketch, represents a continuous reactor in which a component A (feed powder) is converted to product component B as the mixture in process traverses the reactor. Conversion is brought about by reaction with the fluidizing gas. The reaction is:



There will be a concentration gradient of A through the bed because of the reaction (1) taking place. The powder mixture, however, is in constant agitation, and the agitation causes mixing of powder more advanced in conversion with powder of lesser conversion. Consequently, the gradient due to reaction (1) is "flattened" and the concentration of A in product rises undesirably as a consequence. The effect is more pronounced as agitation (fluidization) becomes more vigorous.

An elementary analysis of the effect of mixing is made as an aid in process design. The case developed is for a bed of constant density and a first order reaction. Should results be desired for more complicated situations, machine computations could be made.

MECHANICS OF MIXING

In the absence of agitation, the transfer current* of A at a section a distance s_0 from the bottom would be given by:

$$G_0 c_0 = u_0 \rho_0 c_0 \quad (2)$$

where

G_0 = transfer current up the bed, lb/ft²/hr

c_0 = concentration of A in the mixture

u_0 = bulk velocity of the powder up the bed, ft/hr

ρ_0 = bulk density of the fluidized bed, lb/ft³

Agitation, however, will introduce a current γ_0 of mixture up the bed and an equal current $-\gamma_0$ down the bed. The net current of mixture with or without agitation is always $G_0 = G_0 + \gamma_0 - \gamma_0$.

However, the upward flow of A will change with agitation because the downward flow $-\gamma_0$ will be richer in A (lower than c_0) than is the upward flow γ_0 (higher than c_0).

Considering the upward transfer of A by agitation, assume:

- a. That of the upward transfer current of mixture, a fraction, fd s, originates from an element of bed a distance s below s_0 ; and that the mixture γ_0 fd s is at a concentration c , which is that found at element s .
- b. That a negligible fraction of γ_0 originates below a distance $s_0 - \lambda$.

On this basis, the upward agitation current of A at s_0

$$= \gamma_0 \int_{s_0 - \lambda}^{s_0} c f d s. \quad (3)$$

Likewise, the downward agitation current of A at s_0

$$= -\gamma_0 \int_{s_0}^{s_0 + \lambda} c f d s. \quad (4)$$

* Current implies unit of flow per unit area, lb/ft²-hr

From (3) and (4) the net upward agitation current of A at s_0

$$= \gamma_0 \left(\int_{s_0-\lambda}^{s_0} c f ds - \int_{s_0}^{s_0+\lambda} c f ds \right). \quad (5)$$

Expanding c in a Taylor series about c_0 -

$$c = c_0 + \left(\frac{dc}{ds} \right)_0 (s - s_0) + 1/2 \left(\frac{d^2c}{ds^2} \right)_0 (s - s_0)^2 + \dots \quad (6)$$

From (5) and (6)

(net upward agitation current of A at s_0) = ---->

$$\begin{aligned} \text{---->} &= \gamma_0 \left[c_0 \left(\int_{s_0-\lambda}^{s_0} f ds - \int_{s_0}^{s_0+\lambda} f ds \right) + \left(\frac{dc}{ds} \right)_0 \left(\int_{s_0-\lambda}^{s_0} f(s - s_0) ds \right. \right. \\ &\text{---->} \left. \left. - \int_{s_0}^{s_0+\lambda} f(s - s_0) ds \right) + 1/2 \left(\frac{d^2c}{ds^2} \right)_0 \left(\int_{s_0-\lambda}^{s_0} f(s - s_0)^2 ds \right) + \dots \right] \\ &\text{---->} \end{aligned} \quad (7)$$

Assuming that f is symmetrical about s_0 , all odd terms in the brackets vanish and

(net upward agitation current of A at s_0) ---->

$$= \text{---->} = -2 \gamma_0 \int_{s_0}^{s_0+\lambda} f(s - s_0) ds \left(\frac{dc}{ds} \right)_0$$

----> third and higher order terms. (8)

Now define a mixing term M_0 at s_0 by

$$M_0 \rho_0 = 2 \gamma_0 \int_{s_0}^{s_0+\lambda} f(s - s_0) ds. \quad (9)$$

The units of M_0 are velocity times length (ft^2/hr) which are the same as diffusivity units. By (9), (8) can be rewritten

Net upward agitation current of A at s_0) --->

$$\text{--->} -M_0 \rho_0 \left(\frac{dc}{ds} \right)_0 + \text{third and higher terms.} \quad (10)$$

The total transfer of A up the bed at s_0 is the sum of the bulk transfer and the net transfer resulting from agitation. From (2) and (10) this is

(total upward current of A at s_0) = --->

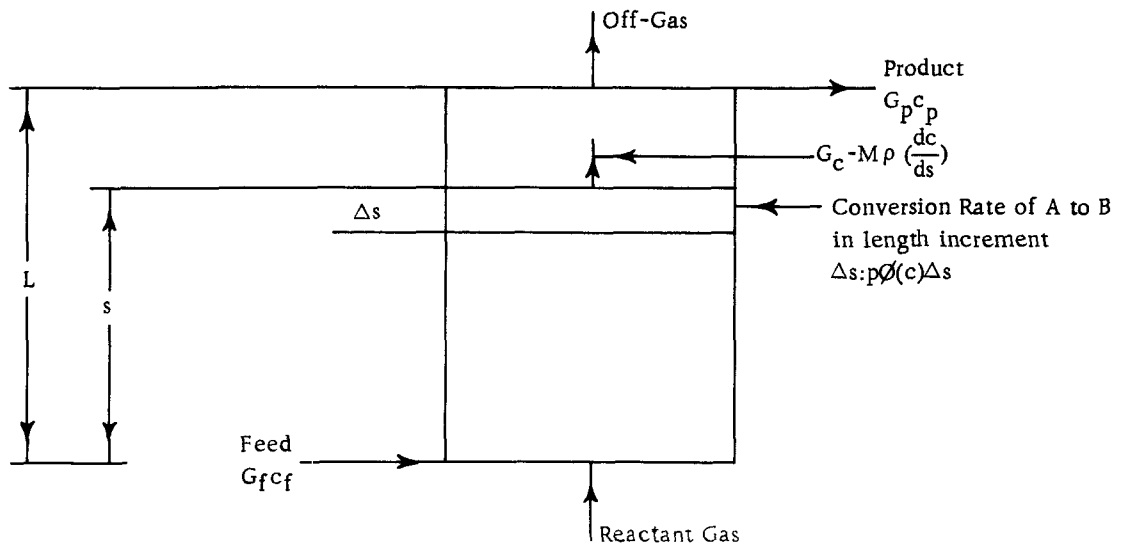
$$\text{--->} G_0 c_0 - M_0 \rho_0 \left(\frac{dc}{ds} \right)_0 + \text{higher order terms.} \quad (11)$$

The higher order terms in (11) contain λ^n , $n \geq 3$. Provided the mixing limit λ is small relative to the length of the bed, the higher order terms can be neglected. The assumption is made then that the agitation is not so violent, as measured by the mixing limit λ , that the higher terms must be carried.

Equation (11) included the effect of mixing on the transfer of component A and will be used to determine the effect of mixing on product concentration. It is interesting to note that (11) above could be stated directly from analogy to presently developed mixing mechanics in gaseous diffusion. However, the entire development is helpful in understanding the theory.

PRODUCTIVITY EQUATIONS FOR A FLUIDIZED BED

Consider the fluidized bed below:



and employ a material balance over the length of bed s , shown up from the feed point.

$$Gc - M\rho \left(\frac{dc}{ds} \right) + \int_0^s \rho \phi(c) ds = G_F c_F \quad (12)$$

Differentiating (12),

$$\frac{d}{ds} \left[Gc - M\rho \left(\frac{dc}{ds} \right) \right] + \rho \phi(c) = 0 \quad (13)$$

which c must satisfy.

The boundary conditions for (13) are obtained by evaluating (12) at $s = 0$ and $s = L$. At $s = 0$

$$\left[Gc - M\rho \left(\frac{dc}{ds} \right) \right]_{s=0} = G_F c_F \quad (14)$$

and at $s = L$

$$\left[Gc - M\rho \left(\frac{dc}{ds} \right) \right]_{s=L} + \int_0^L \rho \phi(c) ds = G_F c_F \quad (15)$$

At $s = h$, $Gc = G_p c_p$ and from the over-all balance

$$G_p c_p + \int_0^L \rho \phi(c) ds = G_F c_F \quad (16)$$

then

$$\left(\frac{dc}{ds} \right)_{s=L} = 0 \quad (17)$$

The boundary equations then are (14) and (17) and the general productivity equation is (13).

CONSTANT MIXING IN A CONSTANT DENSITY BED WHILE A FIRST ORDER REACTION IS UNDERWAY

The results of the previous section are applied to a fluidized bed of constant

density in which is occurring a first order reaction. Referring to the previous sketch (page 62)

$$G_F = G_P = G = V\rho$$

and

$$\rho\phi(c) = \rho kc$$

where

K = first order rate constant,

let

M = mixing constant, $ft^2\text{-hr}^{-1}$

The productivity equations (13), (14), and (17) then are:

$$\left\{ \begin{array}{l} M \frac{d^2c}{ds^2} - \mu \frac{dc}{ds} - Kc = 0 \\ \mu c_F = \left(\mu c - M \frac{dc}{ds} \right)_{s=0} \\ \left(\frac{dc}{ds} \right)_{s=L} = 0 \end{array} \right. \quad \begin{array}{l} (18) \\ (19) \\ (20) \end{array}$$

It is convenient to normalize s by dividing by the bed depth L and letting $\sigma = s/L$.

Two new dimensionless parameters will also be employed:

$R = \frac{KL}{u}$, which is characterized by the reaction rate,

$m = \frac{M}{uL}$, which is characterized by the mixing.

Now (18), (19), and (20) become

$$\begin{cases} m \frac{d^2 c}{d\sigma^2} - \frac{dc}{d\sigma} - Rc = 0 \end{cases} \quad (21)$$

$$\begin{cases} c_F = \left(c - m \frac{dc}{d\sigma} \right)_{\sigma} = 0 \end{cases} \quad (22)$$

$$\begin{cases} \left(\frac{dc}{d\sigma} \right)_{\sigma=1} = 0 \end{cases} \quad (23)$$

The solution of (21) is

$$c = c_1 e^{\alpha\sigma} + c_2 e^{\beta\sigma} \quad (24)$$

where

$$\alpha = \frac{1 + (1 + 4mR)^{1/2}}{2m} \quad (25)$$

$$\beta = \frac{1 - (1 + 4mR)^{1/2}}{2m} \quad (26)$$

c_1, c_2 are constants which can be developed from (22) and (23). Thus

$$\begin{cases} c_F = (1 - m\alpha)c_1 + (1 - m\beta)c_2 \\ 0 = \alpha e^{\alpha} c_1 + \beta e^{\beta} c_2 \end{cases} \quad (27)$$

from which

$$\begin{aligned} c_1 &= c_F \beta e^{\beta} / \left[\beta e^{\beta} (1 - m\alpha) - \alpha e^{\alpha} (1 - m\beta) \right] \\ c_2 &= -c_F \beta e^{\beta} / \left[\beta e^{\beta} (1 - m\alpha) - \alpha e^{\alpha} (1 - m\beta) \right] \end{aligned} \quad (28)$$

Substituting (28) into (24)

$$\frac{c}{c_F} = \frac{\alpha e^{\alpha} e^{\beta\sigma} - \beta e^{\beta} e^{\alpha\sigma}}{\alpha e^{\alpha} (1 - m\beta) - \beta e^{\beta} (1 - m\alpha)} \quad (29)$$

From (25) and (26) it can be shown that

$$1 - m\alpha = m\beta \quad \text{and} \quad 1 - m\beta = m\alpha$$

Substituting in (29) and re-arranging, (29) becomes

$$\frac{c}{c_F} = \frac{m\alpha e^{-(1-\sigma)\beta} - m\beta e^{-(1-\sigma)\alpha}}{(m\alpha)^2 e^{-\beta} - (m\beta)^2 e^{-\alpha}} \quad (30)$$

This then is the concentration gradient in the bed considered. The concentration of feed in product is shown by (30) when $\sigma = 1$.

$$\frac{c_P}{c_F} = \frac{m\alpha - m\beta}{(m\alpha)^2 e^{-\beta} - (m\beta)^2 e^{-\alpha}} \quad (31)$$

Substituting from (25) and (26) into (31)

$$1 - c = \frac{c_P}{c_F} = \frac{4H}{\left[(H+1)^2 e^{\left(\frac{H-1}{2m}\right)} \right] - \left[(H-1)^2 e^{\left(\frac{H+1}{2m}\right)} \right]} \quad (32)$$

where

$$H = (1 + 4mR)^{1/2}$$

and, as before

$$R = KL/u$$

$$m = M/uL$$

This (32) is the expression for product concentration in the fluid bed with simultaneous mixing and reaction.

Two further cases are of interest in connection with (32) above, the cases of no mixing and of extremely violent or instantaneous mixing (no gradient) are taken up. Equation (32) is re-written:

$$\begin{aligned}
 J_2 \rightarrow & \frac{1}{1 - (1 - 1/H)^4 (1 - H/m)} \rightarrow \frac{1}{1 - (1 - 4/H)(1 - 4/m)} \rightarrow \frac{1}{H/m + 4/H} \\
 & \dots \rightarrow
 \end{aligned}
 \tag{36}$$

From (35) and (36)

$$\frac{c_P}{c_F} \rightarrow \frac{4/H}{H/m + 4/H} = \frac{1}{1 + H^2/4m}$$

but

$$\frac{H^2}{4m} = R + 1/m$$

and

$$m = \infty$$

thus,

$$\frac{c_P}{c_F} = \frac{1}{1 + R} = \frac{1}{1 + K L/u}
 \tag{37}$$

APPENDIX D

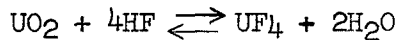
DESIGN OF A COUNTERCURRENT FLUID BED CONTACTOR
BASED ON CONSIDERATION OF AN OVER-ALL DRIVING FORCE

The reagent economy in a fluid bed contactor is related to reactor geometry and over all transport. Given the over-all transport at a given temperature for the maximum available driving force $(1 - y^*)$, the transport for any practical problem is taken as being proportional to the value $(y - y^*)$. This maximum transport is taken as that determined by standard thermogravimetric procedure.

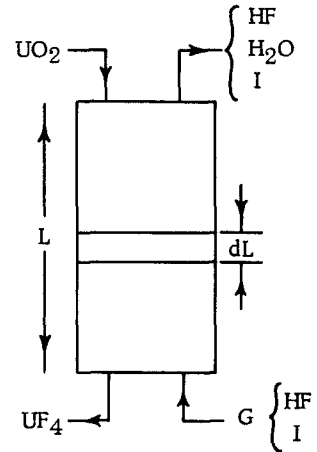
Consider the figure below and assume that

$$J = k(y - y^*) \quad (1)$$

$$J^* = Rk(y - y^*)SdL \quad (2)$$



$$F \frac{dx}{dL} = \frac{SRk(y - y^*)}{I} \quad (3)$$



Let $R = r\rho x$ and combining with (2) and (3) the following integral can be formed:

$$\int_b^t \frac{dx}{x(y - y^*)} = \int_b^t \frac{Sr\rho k}{4F} dL = \frac{Sr\rho k}{4F} L$$

$$L = \frac{4F}{Sr\rho k} \int_{x_b}^{x_t} \frac{dx}{x(y - y^*)} \quad (4)$$

The material balance is

$$4F(x - x_b) = \text{mols HF consumed}$$

$$2F(x - x_b) = \text{mols H}_2\text{O formed}$$

$$\text{mol fraction HF, } y = \frac{Gy_b - 4F(x - x_b)}{Gy_b - 4F(x - x_b) + 2F(x - x_b) + I} \quad (5)$$

$$y = \frac{Gy_b - 4F(x - x_b)}{Gy_b - 2F(x - x_b) + I}$$

Substituting (5) into (4) one obtains

$$\begin{aligned}
 L &= \frac{4F}{Sr\rho k} \int_{x_b}^{x_t} \frac{dx}{x \left(\frac{Gy_b - 4F(x - x_b)}{Gy_b - 2F(x - x_b) + I} - y^* \right)} \\
 &= \frac{4F}{Sr\rho k} \int_{x_b}^{x_t} \frac{[(Gy_b + 2Fx_b + I) - 2Fx] dx}{x [Gy_b + 4Fx_b - 4Fx - y^*(Gy_b + 2Fx_b + I - 2Fx)]} \quad (6)
 \end{aligned}$$

(6) above can be integrated as shown:

Let

$$A = Gy_b + 2Fx_b + I$$

$$B = +2F$$

$$C = Gy_b + 4Fx_b - y^*(Gy_b + 2Fx_b + I)$$

$$D = 4F - 2Fy^* = 2F(2 - y^*)$$

$$L = \frac{4F}{Sr\rho k} \int_{x_b}^{x_t} \frac{(A - Bx)dx}{x(C - Dx)} \quad (7)$$

$$\int_{x_b}^{x_t} \frac{(A - Bx)dx}{x(C - Dx)} = A \int_{x_b}^{x_t} \frac{dx}{x(C - Dx)} - B \int_{x_b}^{x_t} \frac{dx}{C - Dx}$$

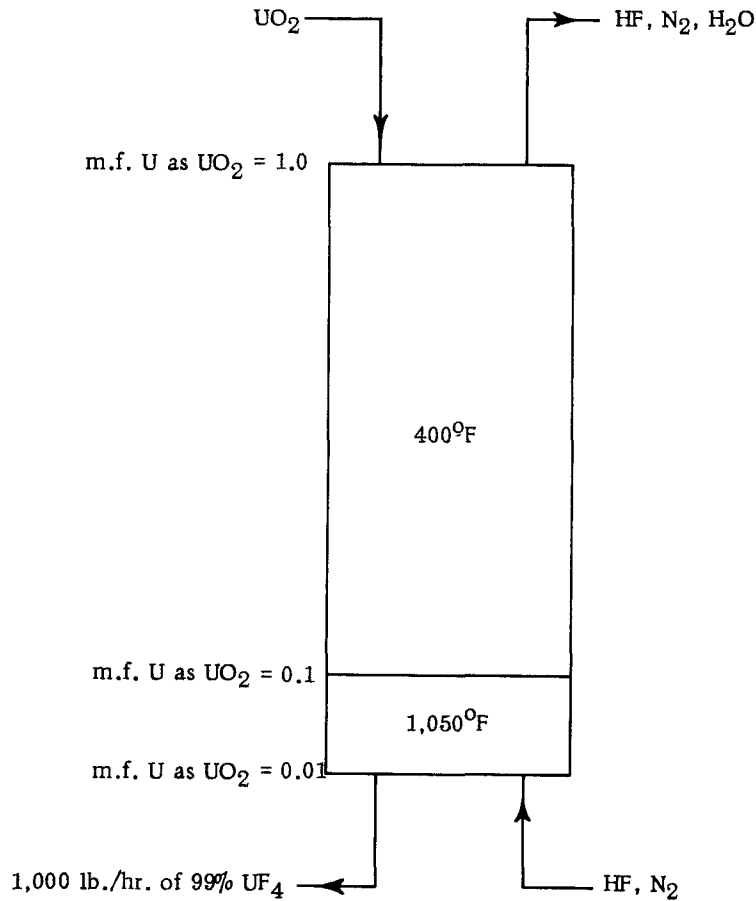
$$= -\frac{A}{C} \ln \left(\frac{C - Dx}{x} \right) + \frac{B}{D} \ln(C - Dx) \Bigg|_{x_b}^{x_t}$$

$$= \frac{A}{C} \ln \left(\frac{C - Dx_b}{x_b} \right) \frac{x_t}{C - Dx_t} + \frac{B}{D} \ln \left(\frac{C - Dx_t}{C - Dx_b} \right) \quad (8)$$

APPENDIX E

SAMPLE DESIGN PROBLEM

One thousand pounds per hour of UF_4 are to be converted in a fluidized bed from a feed consisting of 20-35 mesh, 100% UO_2 . The column is to have two temperature zones, 400° and $1,050^\circ$ F and the UF_4 product purity must be a minimum of 99%. Ninety percent of the conversion is to occur in the low temperature zone and 95% of the HF must be utilized. These conditions are pictorially shown in the sketch below. The entering gas contains 10% nitrogen to afford a means of control and the fluidizing velocity for material in the 20-35 mesh particle size range is 2-3.5 feet per second.



The following data are available:

Temp $^\circ\text{F}$	Rate Constant hr^{-1}	Mol Fraction HF in Equilibrium with UF_4
400	4	.0046
1,050	1	.55

The fluidized density of the powder is 0.926 lb mols per ft^3 (250 lb/ft^3). Assume an over-all heat transfer coefficient of 50 $\text{BTU}/\text{hr}\text{-ft}^2\text{-}^\circ\text{F}$, a ΔT of 200° F (see Table IV). The exothermic heat of reaction (3) is 307 BTU/lb

UO₂. Calculate the length and diameter of the column necessary to satisfy these conditions.

1. Minimum column volume as determined solely from the rate constants (piston flow volume).

For a 1st order reaction,

$$-\frac{dc}{c} = kdt$$

or

$$\frac{c}{c_0} = e^{-kt}$$

cool zone

$$k = 1$$

$$\frac{.1}{1} = e^{-t}$$

$$t = 2.3 \text{ hours}$$

hot zone

$$\frac{.01}{1} = e^{-4t}$$

$$t = 0.6 \text{ hours}$$

Total retention time required = 2.9 hours.

$$\text{No. lb mols to be processed per hour} = \frac{1,000 \text{ lb}}{\text{hr}} \times$$

$$\frac{1 \text{ lb mol}}{314 \text{ lb}} \times \frac{1}{.99} = 3.22$$

$$\begin{aligned} \text{Column volume} &= \frac{3.22 \text{ lb mols}}{\text{hr}} \times \frac{\text{ft}^3}{.926 \text{ lb mols}} \times 2.9 \text{ hrs} \\ &= 10.1 \text{ ft}^3 \end{aligned}$$

2. Minimum area resulting from heat transfer considerations.

$$U = 50 \text{ BTU/hr-ft}^2\text{-}^\circ\text{F}$$

$$\Delta T = 200^\circ \text{ F}$$

$$Q = 307 \text{ BTU/lb}$$

$$\begin{aligned} \text{Area} &= \frac{\frac{307 \text{ BTU}}{\text{lb}} \times \frac{270 \text{ lb}}{\text{lb mol}} \times \frac{3.22 \text{ lb mols}}{\text{hr}}}{\frac{50 \text{ BTU}}{\text{hr-ft}^2\text{-}^\circ\text{F}} \times 200^\circ \text{ F}} \\ &= 24.5 \text{ ft}^2 \end{aligned}$$

$$DL = \frac{24.5}{\pi} = 7.81 \text{ ft}^2$$

3. Volume resulting from mixing efficiency considerations.

From equation (3):

$$1 - c = \frac{4H}{(H+1)^2 e^{(H-1)/2m} - (H-1)^2 e^{-(H+1)/2m}}$$

$$H = (1 + 4mR)^{1/2}$$

$$R = KL/u$$

$$m = \frac{M}{uL}$$

or

$$H = \left(1 + \frac{4MK}{u^2}\right)$$

hot zone:

$$M = 0.4$$

$$K = 4.0$$

$$1 - c = 0.1$$

$$H = \frac{1}{u} (u^2 + 6.4)^{1/2}$$

$$2m = \frac{0.8}{uL}$$

cool zone:

$$M = 0.4$$

$$K = 1.0$$

$$1 - c = 0.1$$

$$H = \frac{1}{u} (u^2 + 1.6)^{1/2}$$

$$(H + 1)^2 e^{(H-1)uL/0.8} - (H - 1)^2 e^{-(H+1)uL/0.8} = 40H$$

The solution to this equation can be obtained by an iterative procedure. The simplest procedure is to assume a value of u and substitute it into the above equation. A trial and error procedure will then yield a value of L corresponding to the assumed value of u .

hot zone

<u>u, ft/hr</u>	<u>L, ft</u>	<u>A, ft²</u>	<u>D, ft</u>	<u>V, ft³</u>
0.5	0.66	6.948	2.97	4.55
1.0	0.96	3.470	1.64	3.34
5.0	3.04	.695	.94	2.11
10.0	5.76	.347	.66	2.0
30.0	17.2	.116	.38	2.0
50.0	29.0	.069	.30	2.0
100.0	57.1	.035	.21	2.0

cool zone

<u>u, ft/hr</u>	<u>L, ft</u>	<u>A, ft²</u>	<u>D, ft</u>	<u>V, ft³</u>
0.5	1.93	6.950	2.97	13.41
1.0	2.92	3.470	1.64	10.13

cool zone (Continued)

<u>u, ft/hr</u>	<u>L, ft</u>	<u>A, ft²</u>	<u>D, ft</u>	<u>V, ft³</u>
5.0	11.52	.695	.94	8.0
10.0	23.03	.347	.66	8.0
20.0	46.06	.174	.47	8.0

These results are plotted in Figure XXI.

4. Volume determined on the basis of HF equilibrium considerations.

$$\frac{1000 \text{ lbs UF}_4/\text{hr}}{314 \text{ lbs/lb mol}} = 3.185 \text{ lb mols UF}_4/\text{hr}$$

$$\frac{3.185}{.99} = 3.217 \text{ lb mols UO}_2/\text{hr}$$

$$\text{HF usage} = \frac{3.185 \times 4}{.95} = 13.41 \text{ lb mols/hr}$$

$$\text{N}_2 \text{ flow} = \left(\frac{13.41}{.9} \right) (.1) = 1.49 \text{ lb mols/hr}$$

a. Hot Zone

$$I = 1.49$$

$$G = 14.9$$

$$F = 3.217$$

$$y_b = 0.9$$

$$y^* = 0.55$$

$$x_b = .01$$

$$x_t = .10$$

$$A = 14.96$$

$$B = +6.434$$

a. Hot Zone (Continued)

$$C = 5.312$$

$$D = 9.329$$

The integral then is

$$\begin{aligned} & \frac{14.96}{5.312} \ln \left[\frac{5.312 - 9.329(.01)}{.01} \frac{.10}{5.312 - 9.329(.10)} \right] \\ & + \frac{6.434}{9.329} \ln \frac{5.312 - 9.329(.1)}{5.312 - 9.329(.01)} \\ & = 2.816 \ln [(521.9)(.02284)] - .6897 \ln \frac{4.379}{5.219} \\ & = 2.816 \ln 11.92 - .6897 \ln .8390 \\ & = 6.984 - .121 \\ & = 6.86 \end{aligned}$$

b. Cool Zone

$$\text{Lbs mols HF used up in hot zone} = (3.217)(4)(.1 - .01) = 1.158$$

$$\text{Lbs mols H}_2\text{O formed} = \frac{1.158}{2} = .579$$

$$\text{Gas flow} = 14.9 - .579 = 14.32$$

$$\text{Mol fraction HF} = \frac{13.41 - 1.158}{14.32} = .856$$

$$I = 1.49$$

$$G = 14.32$$

$$F = 3.217$$

$$y_b = .856$$

$$y^* = .0046$$

$$x_b = 0.1$$

$$x_t = 1.0$$

$$A = 14.39$$

$$B = 6.434$$

$$C = 13.48$$

$$D = 12.84$$

The integral then is

$$\frac{14.39}{13.48} \ln \left[\frac{13.48 - 12.84(.1)}{.1} \frac{1}{13.48 - 12.84} \right] + \frac{6.434}{12.84} \ln \frac{13.48 - 12.84}{13.48 - 12.84(.1)}$$

$$= 1.068 \ln [(122)(1.563)] + .5011 \ln \frac{.64}{12.2}$$

$$= 5.61 - 1.476$$

$$= 4.134$$

$$LS = \frac{4(6.86)F}{\rho r k} ; k = \frac{4}{hr} \times \frac{lb \ mol}{r \ ft^2} \times \frac{1}{(1 - .55) \Delta y}$$

$$= \frac{(4)(3.217)(6.86)}{\rho r \ 4 \times \ 4 \ \frac{lb \ mols \ HF}{lb \ mol \ U} \ r(1 - .55)}$$

$$\rho = .926$$

$$LS = \frac{(4)(3.217)(6.86)(.45)}{(.926)(4)(4)}$$

$$= 2.68 \ ft^3 \quad (\text{hot zone})$$

$$\begin{aligned}
 LS &= \frac{(4)(3.217)(4.134)(1.0)}{(.926)(1)(4)} \\
 &= 14.36 \text{ ft}^3 \quad (\text{cool zone})
 \end{aligned}$$

$$\text{Total volume} = 17.04 \text{ ft}^3$$

Determination of column dimensions.

$$\begin{aligned}
 \text{No. lb mols gas leaving} &= 14.9 - 1/2(3.217)(4)(1 - .01) \\
 &= 6.37 \text{ lb mols/hr}
 \end{aligned}$$

$$\begin{aligned}
 \text{Minimum area at top} &= \frac{nRT}{Pv} \\
 &= \frac{(6.37)(10.73)(860)}{(14.7)(2)(3600)} \\
 &= .556 \text{ ft}^2
 \end{aligned}$$

Allowing for ϵ , the void fraction,

$$\text{Area} = \frac{.556}{.45} = 1.24 \text{ ft}^2$$

$$\begin{aligned}
 \text{Diameter} &= \sqrt{(1.273)(1.24)} \\
 &= 1.26 \text{ ft}
 \end{aligned}$$

$$\text{Column Length} = \frac{\text{vol}}{\text{area}} = \frac{17.04}{1.24} = 13.7 \text{ ft}$$

Assume a length of 15.5 ft to allow for the taper; the pressure drop is 1.74 lbs/in²/ft, giving a bottom pressure of 42 psia; the bottom area is then

$$\frac{(14.9)(1510)(10.73)}{(42)(3600)(.45)(3.5)} = 1.01 \text{ ft}^2$$

The diameter is 1.13 ft

It should be observed that the taper (in this case an angle of 0.25°) was preserved by selecting the necessary fluidization velocity.