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ENGINEERING DEVELOPMENT OF
FLUID-BED FLUORIDE VOLATILITY PROCESSES

*Part 4. Fluidized-packed Beds: Studies of
Heat Transfer, Solids-gas Mixing,
and Elutriation*

by

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Chemical Engineering Division

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TABLE OF CONTENTS

	<u>Page</u>
NOMENCLATURE	12
ABSTRACT	13
I. INTRODUCTION.	13
II. BED EXPANSION AND PRESSURE DROP	15
III. SOLIDS MIXING	18
A. Lateral Solids Mixing	18
1. Diffusion Equation for Solids Mixing	19
2. Apparatus and Procedure.	20
3. Experimental Results	21
4. Effect of Size of Spherical Packing	21
5. Effect of Cylindrical Packing	22
6. Effect of Fluidized Particle Size	23
7. Effect of Bed Height for Fluidized and for Fluidized- packed Beds	24
8. Theoretical Models for Solids Mixing.	25
9. Measurement of Average Particle Velocity	29
10. Fraction of Particles Moved by Bubble.	32
11. Absolute Bubble Velocity	34
12. Conclusions.	38
B. Axial Solids Mixing.	39
1. Apparatus and Procedure.	39
2. Theory for Longitudinal Solids Mixing	42
3. Experimental Results	44
C. Vertical Transport of Fines	51
1. Apparatus and Procedure.	51
2. Two-in.-diameter Column	52
3. Three-in.-diameter Brass Column	53
4. Conclusions.	55
IV. HEAT TRANSFER	56
A. Lateral Effective Bed Thermal Conductivity	56
1. Apparatus and Procedure.	57
2. Experimental Results	60
3. Theory of Heat-transfer Mechanism	61

TABLE OF CONTENTS

	<u>Page</u>
B. Axial Effective Thermal Conductivity.	67
1. Apparatus and Procedure.	67
2. Discussion of Results	69
3. Conclusions.	74
C. Heat-transfer Coefficients	74
1. Heat-transfer Coefficients at the Walls of a Fluidized-packed Bed	74
2. Heat-transfer Coefficients from the Surface of Fixed Packing in a Fluidized-packed Bed	77
V. GAS MIXING	80
A. Fluidized Beds.	80
B. Beds of Fixed Packing.	80
C. Theory	81
D. Equipment and Experimental Procedure	82
E. Experimental Results	86
1. Packed Bed	86
2. Fluidized-packed Bed	90
3. Fluidized Bed without Fixed Packing	91
F. Conclusions.	92
VI. ELUTRIATION.	93
A. Equipment and Experimental Conditions	93
B. Elutriation Studies in the 3- and 5-ft Columns	93
C. Elutriation Studies in the 6-ft Column	99
D. Conclusions.	103
VII. CONCLUSIONS.	104
VIII. DESIGN APPLICATIONS	106
A. Direct Fluorination.	106
1. Heat Transfer	106
2. Temperature Drop across Bed and Wall	107
3. Gas Mixing and Elutriation.	111

TABLE OF CONTENTS

	<u>Page</u>
B. Two-zone Fluorination	111
1. Heat Transfer	111
2. Fluorine Injector Design	115
3. Fines Transport.	118
4. Particle Elutriation	120
5. Conclusions for Two-zone Fluorination	122
APPENDIX: Nomograph for Predicting the Range of Fluidizing Gas Velocities	123
ACKNOWLEDGMENTS	125
REFERENCES	126

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Bed Configurations for Study of Fluidization Characteristics of a Fluidized-packed Bed	15
2.	Comparison of Fluidized Bed Expansion for Open-tube Fluidization and Fluidized-packed Beds	16
3.	Comparison of Bed Pressure Drop for Open-tube Fluidization and Fluidized-packed Beds.	17
4.	Sample Concentration Profile	19
5.	Schematic Diagram of Apparatus for Lateral Mixing in a Fluidized-packed Bed	20
6.	Effect of Size of Spherical Packing on Solids Mixing in a Fluidized-packed Bed	22
7.	Mixing Diffusion Coefficients for a Fluidized-packed Bed with Cylindrical Packing	22
8.	Effect of Fluidized Particle Size on Solids Mixing	23
9.	Effect of Fluidized Particle Size Distribution on Solids Mixing	24
10.	Effect of Bed Height on Rate of Solids Mixing in a Fluidized Bed	25
11.	Comparison of Theory and Data for Diffusion of Fluidized Particles in Voids of 1/2-in. Spheres.	27
12.	Fluidized Particle Deflection by Spherical Packing.	28
13.	Dependence of Solids Diffusivity on Fixed Packing Diameter	29
14.	Average Fluidized Particle Velocity	30
15.	Drag on Fluidized Particles	32
16.	Bed Expansion Ratios	35
17.	General Correlation for Solids Mixing in a Fluidized-packed Bed	37
18.	General Correlation of Solids Mixing in a Fluidized-packed Bed Neglecting Relative Bed Expansions	38
19.	Concentration Profile for Longitudinal Solids Mixing in a Fluidized-packed Bed	41
20.	Effect of Gas Velocity on Longitudinal Solids Mixing Based on Theoretical Model	43

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
21.	Effect of Bed Height on Longitudinal Solids Mixing in a Fluidized-packed Bed of -140 +170 Copper-Nickel Shot	45
22.	Effect of Bed Height on Longitudinal Solids Mixing in a Fluidized-packed Bed of -100 +120 Copper-Nickel Shot	45
23.	Effect of Bed Height on Longitudinal Solids Mixing in a Fluidized-packed Bed of -40 +50 Copper-Nickel Shot	46
24.	Effect of Size of Fixed Packing on Longitudinal Solids Mixing of -140 +170 Mesh Particles at 8-in. Bed Height	47
25.	Effect of Fixed Packing Size on Longitudinal Solids Mixing of -100 +120 Mesh Particles at 16-in. Bed Height	48
26.	Effect of Fixed Packing Size on Longitudinal Solids Mixing of -100 +120 Mesh Particles at 8-in. Bed Height	49
27.	Effect of Change in Void Structure of Fixed Packing on Longitudinal Solids Mixing in a Fluidized-packed Bed	50
28.	Nickel Fines Particle Size Analysis by Coulter Counter	51
29.	Comparison of Vertical Fines Transport Rate with Longitudinal Fluidized Particle Mixing Rates	53
30.	Concentration of Fines in Alumina Grain Bed after Various Time Intervals of Fluidized-packed-bed Operation	54
31.	Equilibrium Concentration Profile of Fines in Alumina Grain Resulting from Fluidized-packed-bed Operation	54
32.	Schematic Diagram of Apparatus for Measuring Lateral Thermal Conductivities in a Fluidized-packed Bed	57
33.	Comparison of Solids Mixing Diffusivities with Thermal Diffusivities for Copper-Nickel Fluidized Particles	61
34.	General Correlation for Thermal Diffusivities	65
35.	General Correlation for Thermal Diffusivities Neglecting Density Changes because of Bed Expansion	66
36.	Column for Measuring Longitudinal Thermal Conductivity in a Fluidized-packed Bed	68
37.	Longitudinal Temperature Profile in a Fluidized-packed Bed	70
38.	Longitudinal Thermal Conductivities for Different Fluidized Material	70
39.	Longitudinal Thermal Conductivities for Different Shapes of Packing Material	71

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
40.	Longitudinal Thermal Conductivities for Fluidized-packed Beds, Based on Total Heat Flux	73
41.	Fluidized-packed-bed Heater and Wall Film Coefficient, h , as a Function of Gas Velocity	75
42.	Heater and Wall Heat Transfer Coefficients for Different Sizes of Copper-Nickel Shot Fluidizing in the Voids of 3/8-in. Spherical Packing.	76
43.	Fixed Packing Surface Heat Transfer Coefficients in a Fluidized-packed Bed	78
44.	Heat-transfer Coefficients for Three Individual Pellet Heaters in a Fluidized-packed Bed	79
45.	Hiby and Schümmer Relationship for Sample Concentration and Diffusivity	83
46.	Schematic Diagram of Column Used for Eddy Diffusion Measurements	83
47.	Concentration Profile of Tracer Gas in a Packed Bed	85
48.	Semilog Plot of Concentration as a Function of r^2	85
49.	Peclet Number as a Function of Gas Velocity for Packed Beds	86
50.	Peclet Numbers for Fluidized-packed Bed Packed with 1/2-in. Spheres	90
51.	Peclet Numbers for Fluidized-packed Bed Packed with 1/4-in. Spheres	90
52.	Peclet Numbers for Fluidized-packed Bed Packed with 1/8-in. Spheres	90
53.	Peclet Numbers for Fluidized-packed Beds Packed with 3/8 x 3/8-in. Cylinders	90
54.	Peclet Numbers for Fluidized Bed and for an Empty Column .	92
55.	Comparison of Gas Turbulence in Various Systems	92
56.	Elutriation Rate for 3-ft Column	94
57.	Elutriation Rates for 5-ft Column	95
58.	Correlation of Elutriation Data for a Rate Constant Independent of Fines Concentration (Leva's Model)	96
59.	Effect of Chain Length on Average Drag per Sphere	96

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
60.	Assumed Effect of Fines Concentration in a Fluidized Bed on the Average Drop per Fine Particle.	97
61.	Correlation of Elutriation Data for an Elutriation Rate Constant Depending upon Concentration to the One-half Power	98
62.	Correlation of Elutriation Data for an Elutriation Rate Constant Depending upon Concentration to the First Power . .	98
63.	Elutriation of U_3O_8 Fines from a Mixture of U_3O_8 Fines in Fluidized Alumina, as a Function of Fluidizing Gas Velocity .	99
64.	Elutriation of U_3O_8 Fines from a Mixture of U_3O_8 Fines in a Fluidized Bed of Alumina, as a Function of Pulse Frequency.	100
65.	Elutriation of U_3O_8 Fines from a Mixture of U_3O_8 Fines in a Fluidized Bed of Alumina, as a Function of Pulse Gas Manifold Pressure	100
66.	Elutriation of U_3O_8 Fines from a Mixture of U_3O_8 Fines in Fluidized Alumina, as a Function of UO_2 Pellet Bed Height and Gas Pulse Frequency.	101
67.	Elutriation of U_3O_8 Fines from a Mixture of U_3O_8 Fines in Fluidized Alumina, as a Function of Pulse Gas Manifold Pressure and UO_2 Pellet Bed Height	101
68.	Elutriation of U_3O_8 Fines from a Mixture of U_3O_8 Fines in Fluidized Alumina in a Packed Bed, as a Function of Alumina Particle Size	102
69.	Flow-schematic Diagram for Direct Fluorination of UO_2	106
70.	Temperature Differential between Center of Bed and Wall for Direct Fluorination of UO_2 Pellets.	110
71.	Fluorine Distribution for a Single Injector for Two-zone Fluorination	118
72.	Nomograph for Predicting the Range of Fluidizing Gas Velocities	124

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	Drag Ratios for Different Copper-Nickel Particle Sizes . .	33
II.	Comparison of Bubble Velocities.	35
III.	Calculation of Minimum Fluidization Velocities	37
IV.	Thermal Conductivities and Properties of the Fluidized-packed Beds	59
V.	Fluidized Particle Size Distributions	69
VI.	Experimental Data for 2000 g of No. 070 Glass Beads in a 19-in.-deep Bed of 3/8 x 3/8-in. Brass Cylinders	71
VII.	Experimental Data for 2000 g of No. 120 Glass Beads in a 19-in.-deep Bed of 3/8 x 3/8-in. Brass Cylinders	72
VIII.	Experimental Data for 2000 g of 56/100 Mesh Alumina in a 19-in.-deep Bed of 3/8 x 3/8-in. Brass Cylinders	72
IX.	Experimental Data for 2000 g of 56/100 Mesh Alumina Grain in a 17-in.-deep Bed of 1/2-in.-diam Steel Spheres .	72
X.	Peclet Numbers for 1/2-in. Spherical Packing.	87
XI.	Peclet Numbers for 1/4-in. Spherical Packing	87
XII.	Peclet Numbers for 1/8-in. Spherical Packing	88
XIII.	Peclet Numbers for 3/8-in. Cylindrical Packing	88
XIV.	Peclet Numbers for Open Tube	89
XV.	Peclet Numbers for Various Systems Based on Column Diameter	89
XVI.	Conditions for Elutriation Tests with the 5- and 3-ft Columns	94
XVII.	Heat-transfer Coefficients and Thermal Conductivities for Fluidized-packed Bed of UO ₂ Pellets and 0.2-mm Alumina Grain	108
XVIII.	Comparison of Temperature Differentials Obtained during Fluorination with Those Calculated from Heat-transfer Tests	110
XIX.	Fluorine Efficiencies at Different Heights above Gas Injector	119

NOMENCLATURE

a	Initial column height containing nickel ft	Na	Mass transfer rate, g-mole/(cm ²)(sec)
A	Area sq ft	N _f	Rate of fines transport, lb/(ft ²)(sec)
A _s	Cross-sectional area sq ft	p	Partial pressure, atm
A _r	Radius of injector and sample tube ft	P	Factor for effect of pressure
b	Correction for available volume cu ft	q	Heat flux Btu/(hr)(ft ²)
C	Concentration fractional weight (Sec III and VI)	q'	Rate of heat generation Btu/(ft ³)(hr)
C	Concentration, lb/ft ³ (Sec IV and VIII)	Q	Sensible heat of particle Btu/lb (Sec IV-A)
C	Concentration, lb-moles/ft ³ (Sec V)	Q	Heat flow Btu/hr (Sec IV-B)
C	Drag coefficient (Appendix)	r	Radial coordinate, ft
C _a	Average concentration of tracer in total gas stream, lb-moles/ft ³ (Sec V)	r _b	Radius of bed, ft
C _D	Drag coefficient	R	Expansion ratio (Sec III)
C _D	Drag coefficient for single sphere	R	Gas law constant 82.1 (cm ³)(atm)/(g-mole)(°K) (Sec VIII)
C ₀	Initial concentration fractional weight (Sec III and VI)	R	Radius of column (Sec V)
C ₀	Initial concentration lb/ft ³ (Sec VIII)	R _e	Reynolds number
C ₀	Concentration of tracer in injected gas, lb-mole/ft ³ (Sec V)	R _{emf}	Reynolds number at minimum fluidization
C _f	Fines concentration in fluorination zone lb/ft ³	R _p	Particle radius ft
C _p	Heat capacity, Btu/(lb)(°F)	S	Cross-sectional area, ft ²
C _s	Particle heat capacity Btu/(lb)(°F)	t	Temperature, °F
$\bar{C}$	Average concentration in one half of bed, fractional weight (Sec III)	T	Temperature, °F
$\bar{C}$	Sample concentration lb-mole/ft ³ (Sec V)	T _s	Temperature of surroundings, °F
$\bar{C}_D$	Mean drag coefficient	T ₁	Initial particle temperature °F
d	Diameter of fluidized particle	T ₂	Final particle temperature, °F
D	Solids-mixing diffusivity ft ² /sec	u	Average velocity of fluidized particle ft/sec
D _f	Drag, lb-force	V	Gas velocity (corrected for packing voidage) ft/sec
D _p	Diameter of fixed packing, ft	V _b	Volume of fluidized bed ft ³
D _t	Column diameter, ft	V _B	Absolute bubble velocity, ft/sec
D _v	Molecular diffusivity, ft ² /sec	V _{B 0.0054}	Absolute bubble velocity in bed of 0.0054-in particles ft/sec
E	Eddy diffusivity ft ² /hr	V _{mf}	Minimum fluidization velocity, ft/sec
E _f	Amount of fines elutriated	V _{mf 0.0054}	Minimum fluidization velocity for 0.0054-in particles, ft/sec
E _t	Total diffusivity (eddy plus molecular) ft ² /hr	V ₀	Superficial gas velocity, ft/sec
erf	Error function	V _{0mf}	Minimum Superficial gas velocity, ft/sec
F	Volume of particles moved/volume of gas forming bubbles for 0.0054-in diam reference particle system	V _p	Volume of particles moved by gas flow through bubble, ft ³
F'	Volume of particles moved/volume of gas forming bubbles	V _s	Volume of sphere, ft ³
F _f	Fraction of fines escaping from bed	W	Volumetric gas flow rate, ft ³ /sec
g	Gravitational constant, 32.17 ft/sec ²	W _{mf}	Minimum fluidization volumetric gas flow rate ft ³ /sec
G	Volumetric flow rate, cfm	x	Distance, ft
h	Heat-transfer coefficient, Btu/(hr)(ft ²)(°F)	$\bar{\Delta X}$	Mean free path of particle, ft
H	Half-column width, ft	y	Distance ft
ΔH	Heat of reaction Btu/lb-mole	z	Longitudinal distance, ft
J ₀ (x)	Bessel function	α	Thermal diffusivity, ft ² /sec (Sec IV and VIII)
J ₁ (x)	Bessel function	α'	Thermal diffusivity based on ρ ft ² /sec
k	Thermal conductivity, Btu/(hr)(ft ²)(°F/ft)	α	Angle, radians (Sec III)
k _c	Mass transfer coefficient, cm/sec	α _s	Shape factor for gamma distribution
k _e	Effective bed thermal conductivity, Btu/(hr)(ft ²)(°F/ft)	ε	Fixed packing void fraction
k _g	Thermal conductivity of gas, Btu/(hr)(ft ²)(°F/ft)	ε _B	Fluidized-bed void fraction
k _r	Reaction rate coefficient, g-mole/(cm ²)(hr)	ε _{B 0.0054}	Fluidized-bed void fraction for 0.0054-in particles
K	(F)(V _{mf 0.0054}) (Sec III)	ε _h	Emissivity
K	Ratio of column radius to packing diam, R/D _p (Sec V)	ε ₀	Fluidized-particle void fraction at static conditions
K	Elutriation rate constant (Sec VI)	θ	Time
K	Reaction rate, sec ⁻¹ (Sec VIII)	θ _R	Time to completely react a fine particle
K'	Coefficient for vertical drift (Sec III)	$\bar{\theta}$	Mean particle residence time
K'	A constant involving frictional drag (Sec VI)	λ ₀ = 0, λ ₁ = 3.837, λ ₂ = 7.0156, λ ₃ = 10.1735 etc	
K _E	Elutriation rate constant, C ^{-1/2} min ⁻¹	μ	Gas viscosity, lb/(hr)(ft)
K _s	Correction for space occupied by particles, ft ³	ν	Number of particles
L	Bed height, ft	ρ	Density, lb/ft ³
L ₀	Bed height of static conditions, ft	ρ _B	Fluidized bed density, lb/ft ³
ΔL	Bed expansion, ft	ρ _f	Fluid density, lb/ft ³
M _p	Fluidized particle weight, lb	ρ _s	Density of particle, lb/ft ³
n	Number of collisions	σ	Stefan-Boltzmann constant
N	Mass transfer rate, lb/hr (Sec IV and Sec VIII)	Σ	Surface area of walls, ft ²
N	Mass transfer rate, lb-mole/(ft ²)(hr) (Sec V)	ø	C _e ^{Kz/V₀}

ENGINEERING DEVELOPMENT OF FLUID-BED FLUORIDE VOLATILITY PROCESSES

Part 4. Fluidized-packed Beds: Studies of Heat Transfer, Solids-Gas Mixing, and Elutriation

by

John D. Gabor and William J. Mechem

ABSTRACT

A system involving the fluidization of small particles in the voids of a packed bed of larger particles has been referred to as a "fluidized-packed" bed. Because of the movement of the fluidized particles, improvement of transport properties for the fluidized-packed bed is attained in comparison to a packed bed. The fluidized-packed bed has found application in the development of a fluoride volatility process for the recovery of fissionable values from spent uranium dioxide reactor fuels. In this process, a highly exothermic fluorination is carried out. The uranium dioxide pellets are too large to be fluidized directly, but the addition of inert fluidized, alumina grain has improved heat and mass transfer from the reacting surfaces of the pellets.

Various properties of fluidized-packed beds have been studied from a basic point of view: bed expansion and pressure drop, solids mixing, gas mixing, effective bed thermal conductivity, heat-transfer coefficients at the column wall and fixed packing surface, and elutriation. Data and theoretical discussions of mechanisms are presented. In addition, the empirical and theoretical results are applied to process design and, where possible, compared with pilot-plant data.

I. INTRODUCTION

Fluidization techniques are well-known for their application to solid-gas contactors where high production rates and good heat transfer are required. In the area of reactor fuel processing, various applications of fluidization have been studied at Argonne National Laboratory since 1953. Fluidized-bed techniques have been applied to a commercial process for

production of uranium hexafluoride feed material from ore concentrates.⁽³⁹⁾ In this process, the uranium oxide feed was briquetted, crushed, and screened to provide a particle size suitable for fluidization.

In a process for recovery of uranium from spent reactor fuels of the enriched uranium-zirconium alloy type, the fuel assembly (of a multiple form) was immersed in a fluid bed of inert particles to aid in the removal of heat generated by the chlorination of the fuel plates.⁽²⁷⁾

The present study of fluidized-packed beds derives from development work on a fluidized-bed fluoride volatility process applied to power reactor fuel of the low-enrichment uranium dioxide type.⁽³⁴⁾ This dense oxide fuel is typically in the form of rods or pellets of about 1/2-in. diameter. Because the fuel as received is too massive to be fluidized directly, the fuel segments were immersed in a fluidized bed of inert particles. A possible configuration of the charge to the fluorinator in this process is a packed bed, of 1/2 x 1/2-in. cylindrical pellets, which forms a network of voids in which the granular material is fluidized. Such a system of particles fluidized in the voids of a bed of nonfluidized larger bodies is referred to as a fluidized-packed bed.

Although extensive work has been done on the heat transfer and other aspects of fluidized beds and packed beds separately, there have been few reports on the combination fluidized-packed bed. The most pertinent reports are cited in the following paragraphs.

Matheson⁽³²⁾ has a patent for filling a fluidized-bed reactor with Raschig rings or Berl saddle packing, to disperse bubbles by providing a tortuous path for the gas. Sutherland *et al.*⁽⁴⁸⁾ reported minimum fluidization velocities, pressure drops, bed expansion, and heat transfer in fluidized-packed beds. Zenz and Othmer⁽⁵⁹⁾ mention the use of a bed of large spheres to inhibit the down flow of fluidized particles in a multi-stage fluidized-bed reactor, and further development of this principle has been described by Williamson.⁽⁵⁵⁾ Ziegler and Brazelton⁽⁶⁰⁾ have discussed the mechanism of radial heat transfer in fluidized-packed beds and published data from work performed at Argonne National Laboratory in cooperation with Northwestern University. Baskakov and Vershinina⁽⁶⁾ measured heat-transfer coefficients between the packing surfaces and the fluidized bed in the interstices of the packed bed.

Because of the limited data available, studies were undertaken in support of the development of fluidized-bed fluoride volatility processes at Argonne National Laboratory. Investigations were made of bed expansion, pressure drop, heat transfer, solids mixing, gas mixing, and fines elutriation. These investigations showed that heat transfer was markedly better for fluidized-packed beds than for comparable packed beds. On the other hand, the presence of packing results in lower rates of heat transfer

in the fluidized-packed bed than the rates for usual mode of fluidization. Eddy diffusivities of the gas in the radial direction in fluidized-packed beds are found to be nearly the same as those for beds with the same type of packing, but without the fluidized material. Measured rates of elutriation of fine particles from a fluidized-packed bed are less than those from a comparable fluidized bed (no packing). Rates of mixing of solid particles obtained in these experiments show values comparable to those obtained from heat-transfer rates when interpreted according to the common mechanism of solids mixing involved in the two processes.

The theoretical models and the data correlations developed in these investigations are applied to the analysis of some typical processing conditions in pilot-plant tests of fluid-bed fluoride volatility processing.

II. BED EXPANSION AND PRESSURE DROP

Tests with fluidized-packed-bed systems were made in a glass column to observe the fluidization characteristics and to judge qualitatively the feasibility of the fluidized-packed-bed system for use as a chemical reactor. Bed expansion and pressure drop measurements were made as well as visual observation of the fluidization quality. These tests were not extensive, but were intended to determine whether fluidized particle movement in the constrictive voids of the fixed packing would be sufficient for a process reactor.

Copper shot ($-140 +200$ mesh) was fluidized in a bed of nickel balls ranging from $1/8$ to $3/8$ in. in diameter. The column was a 3-in.-diameter glass tube with a porous metal disk as the gas distributor. Air was used as the fluidizing gas.

Three systems were studied as shown in Figure 1. Case A was a normal (no fixed packing) fluidized bed of copper shot, approximately 6 in. deep at static conditions; Case B was a 6-in. bed of nickel balls, with the voids filled with copper shot; and Case C was an 11-in. bed of nickel balls, with the voids filled to a 6-in. depth with copper shot. Bed expansion as a function of air velocity for the three systems is plotted in Figure 2. The indicated gas velocities are superficial, i.e., uncorrected for fixed packing voidage. The bed expanded linearly with increased air velocity for the case of the fluidized bed (no fixed packing). For the 6-in. fluidized-packed bed (Case B), the amount of expansion was also a linear function of air velocity, but the rate of expansion was less than for the usual fluidized-bed case. The total charge of fluidized

Figure 1
BED CONFIGURATIONS FOR STUDY
OF FLUIDIZATION CHARACTERISTICS
OF A FLUIDIZED-PACKED BED

particles for Case B was less than for Case A because of the space occupied by the fixed packing. During fluidization, the bed consisted of particles fluidized in the voids of the static bed of nickel balls, and a layer of particles that had expanded above the static bed. As a result, the amount of bed expansion, which was due chiefly to the expansion of the fluidized particle layer above the bed of nickel balls, was less than for the conventional fluidized-bed case. The increase in bed-expansion ratio for the 11-in. bed (Case C) was greater than that for the fluidized bed, except where the fluidized particles expanded above the fixed bed, at which point there is a break in the curve. Since the gas velocities indicated on Figure 2 are superficial, correction for the fixed packing void fraction in the case of the fluidized-packed bed would result in higher fluidization gas velocities. The bed expansion would then be greater because of the higher gas velocity.

Figure 2
COMPARISON OF FLUIDIZED
BED EXPANSION FOR OPEN-
TUBE FLUIDIZATION AND
FLUIDIZED-PACKED BEDS

Pressure drops across the beds are plotted in Figure 3. A mercury manometer was used to indicate bed pressures. At the higher velocities, particularly for the nonpacked fluidized bed, there was oscillation of up to 25 percent from the mean in the readings, resulting in less precise measurements. For the fluidized bed, the differential pressure across the bed is nearly constant and approximately equivalent to the weight of the material per unit cross-sectional area. For the system in which the fluidized bed is expanded above the static bed, the pressure drop across the bed exhibits a decrease with increasing gas flow. This decrease is in accordance with the observation that a given weight of fluid

Figure 3

COMPARISON OF BED PRESSURE DROP FOR
OPEN-TUBE FLUIDIZATION AND
FLUIDIZED-PACKED BEDS

will exert a greater static pressure in a small-diameter tube than in a large-diameter tube; thus,

$$\text{pressure} = \frac{\text{weight}}{\text{cross-sectional area.}}$$

The fluidized phase, which exhibits a hydrostatic behavior similar to that of a true liquid, is initially confined to the voids in the static bed. But with increasing air velocity, the fluidized material is expanded into the large open tube. A given weight of material then exerts a lower head in the larger tube section than in the fixed bed. Thus, progressive expansion leads to a progressive loss in total static head.

For the 11-in. bed, there is a slight decrease in bed pressure drop with increasing gas velocity until the fluidized particles expand

to the top of the fixed bed. Then, in the region where the particles expand above the fixed bed, the pressure drop decreases more rapidly.

The above data show the following marked changes in fluidization characteristics for a fluidized-packed bed, compared to normal open tube fluidization:

1. Greater bed expansion at the same superficial gas velocity.
2. Decrease in bed pressure drop as fluidized bed expands above bed of fixed packing.

Although these differences exist, direct visual observation indicated that satisfactory fluidization did exist in the voids of the fixed packing. The fluidization for the fluidized-packed bed was quite smooth, and slugging did not occur at superficial gas velocities that would cause violent slugging in a fluidized bed. However, the solids motion was not as rapid in the fluidized-packed bed as in a fluidized bed. Subsequent heat transfer and solids mixing tests gave quantitative measures of the reduced solids movement, as will be discussed in Sec. III.

Mock-up tests indicated that minimum fluidization velocities for fluidized-packed beds were essentially the same as for a fluidized bed, if the superficial gas velocity were corrected for the void fraction of the fixed packing. Sutherland *et al.*⁽⁴⁸⁾ found that the maximum fluidization velocities, corrected for fixed packing void fraction for a fluidized-packed bed, could be up to a factor of two greater than the minimum velocity for open-tube fluidization.

III. SOLIDS MIXING

An understanding of the motion of fluidized particles is basic to the understanding of the properties of fluidized-packed beds. One advantage of a fluidized bed, compared to a packed bed, is the movement of the fluidized particles. The moving particles in a fluidized bed absorb and transport essentially all of the heat,^(23,58,61) whereas heat transfer in the packed bed is primarily by the gas eddies. Because the volumetric heat capacity of solid particles is much greater than that of gas, the heat content of the particles is greater. Hence, heat transport by the fluidized particles is greater.

In oxidations of uranium dioxide pellets, the fine uranosic oxide (U_3O_8) particles were removed rapidly from the surface of the pellets when an inert material was fluidized in the voids of the uranium dioxide pellet bed.⁽¹⁸⁾ The fluidized particle movement apparently provided an abrasive action which reduced the adherence of fines to the pellet surfaces.

It would be expected that solids mixing would be less in a fluidized-packed bed than for the usual fluidized bed because of baffling by the fixed packing. The literature indicates little concerning the degree to which the solids movement would be hindered by the fixed packing. Bart,⁽⁵⁾ Massimilla and Bracale,⁽³⁰⁾ Hayakawa *et al.*,⁽²²⁾ Talmor and Benenati⁽⁴⁹⁾ obtained quantitative measurements for longitudinal solids mixing in fluidized beds. Brötz⁽⁹⁾ worked on lateral solids mixing of fluidized beds. While these references are of interest for analysis and technique, they give no indication of the effect that fixed packing would have on the mixing rates. Measurements were therefore made of rates of lateral and longitudinal solids mixing in fluidized-packed beds. The mechanism of fluidized particle mixing was theorized, and empirical data were compared with theory. A few measurements were also made of rates of vertical fines movement.

A. Lateral Solids Mixing

In the study of lateral solids mixing, the experimental method of measuring the rate of solids mixing was patterned after that of Brötz.⁽⁹⁾ The general method is based on the use of two solids that are similar with respect to the physical properties determinative of the mixing behavior, but are different on the basis of some property usable for analysis of composition. The analytical property employed by Brötz was color, but for the present study the differences in ferromagnetic properties of nickel and copper were used. Since nickel and copper have the same density (8.9 g/cc), they should exhibit identical fluidizing characteristics for the same particle size and shape.

Initially, the pure copper particles are separated by a centrally-located partition in a rectangular cell from the pure nickel particles, and

the particles are fluidized in both sections. Then the partition is removed, and mixing occurs on each side of the center line. After the particles are allowed to mix for a desired time interval, the mixing is arrested by shutting off the fluidizing gas. Bed samples of the particles are taken at known distances from the center line of mixing, and analyzed for weight concentration by magnetic separation. The concentration at each known distance allows a concentration profile to be plotted. A typical set of experimental points is plotted in Figure 4. The location of the center line is shown at the distance $x = 0$.

Figure 4
SAMPLE CONCENTRATION PROFILE

108-6187 Rev.

1. Diffusion Equation for Solids Mixing

A diffusion coefficient for the bed of fluidized solids was used to characterize the rate of solids mixing. The usual differential equation for diffusion in one dimension is

$$\frac{\partial C}{\partial \theta} = D \frac{\partial^2 C}{\partial x^2}. \quad (1)$$

The solution of this equation for the described experimental conditions is given by Sherwood and Reed:⁽⁴⁶⁾

$$C = 1/2 + \frac{2}{\pi} \left\{ \exp \left[- \left(\frac{\pi}{2} \right)^2 \frac{D\theta}{H^2} \right] \sin \frac{\pi x}{2H} + (1/3) \exp \left[- 9 \left(\frac{\pi}{2} \right)^2 \frac{D\theta}{H^2} \right] \sin \frac{3\pi x}{2H} \right. \\ \left. + (1/5) \exp \left[- 25 \left(\frac{\pi}{2} \right)^2 \frac{D\theta}{H^2} \right] \sin \frac{5\pi x}{2H} + \dots \right\}. \quad (2)$$

In Equation 2, values of the concentration, C , are related to values of diffusion distance, x , in terms of the diffusivity, D , one-half the column width, H , and the time, θ , of mixing. Thus the diffusion coefficient, D , was found from the value of the parameter which fit the experimentally measured profile of C as a function of x . Good fits were obtained; a typical one is shown in Figure 4. Fits were made visually with an estimated 10 to 20% accuracy in D (see Figure 19).

2. Apparatus and Procedure

A schematic diagram of the apparatus is shown in Figure 5. Nitrogen was the fluidizing gas and was distributed by a porous metal plate. A rotameter measured gas flow rates. Sampling was accomplished by a horizontal row of $1\frac{1}{2}$ -in.-long, fixed, hypodermic needles, $\frac{3}{4}$ in. above the bottom, spaced 1 in. apart. Also, samples were taken by a 9-in. long needle that was injected into the bed to the desired position. The concentration profile was invariant with respect to height because of high vertical mixing rates.

Figure 5

SCHEMATIC DIAGRAM OF APPARATUS FOR LATERAL MIXING IN A FLUIDIZED-PACKED BED

108-6193 Rev. 2

To prevent static effects, the fluidizing gas was humidified by passage through a 1-ft packed column, partially filled with water.

The fluidizing material consisted of copper and nickel shot of -40 +50, -60 +70, -100 +120, -120 +200, and -140 +170 mesh sizes. Fixed

packing consisted of 1/2-, 3/8-, 1/4-, and 3/16-in.-diameter steel spheres and 1/2 x 1/2-, 3/8 x 3/8-, and 1/4 x 1/4-in. brass cylinders. A set of runs was also made without fixed packing, i.e., with normal open-tube fluidization.

A rubber partition was inserted in the center of the column. One half of the column was charged with fixed packing and nickel shot, and the other half was charged with fixed packing and copper shot. A needle valve was preset to give the desired flow rate. Then the partition was removed, and fluidization over a measured time interval was effected by opening and closing a quick-opening toggle valve in the gas line. Samples of the mixed particulate solids were withdrawn by means of the hypodermic needles. The sample mixtures were spread out on a flat surface, and the nickel particles were removed by passing a small magnet over the surface. Direct weighing of the separated fractions was made to establish sample compositions.

The run time varied from about 0.5 to 20 min depending on the gas velocity and the fluidized particle size. Smooth fluidization was attained in the order of about a second after introduction of the fluidizing gas. It was, therefore, expected that initial startup disturbances would have negligible effect on the data.

3. Experimental Results

The mixing experiments performed resulted in values of the diffusion coefficient, D , as a function of gas velocities for various fluidized-packed-bed systems. Variations of D were investigated for different sizes of spherical and of cylindrical packing, as well as different sizes and heights of fluidized material. Reference cases for fluidization without packing are also presented.

4. Effect of Size of Spherical Packing

A series of runs was made with packing consisting of 1/2-, 3/8-, 1/4-, and 3/16-in.-diameter spheres and 0.0038-in.-diameter copper and nickel shot. The data for the several packings are plotted as D versus average gas velocity in Figure 6. The diffusion coefficients increased with packing size over the range tested. Fluidization was also attempted in a bed of 1/8-in. spheres, but the particle motion was greatly inhibited and channeling occurred. For open-tube fluidization (no packing), mixing diffusion coefficients are about three and a half times higher than for 1/2-in. packing at the same gas velocities. The fluidized-bed data were for an expanded bed height of approximately 1 in. The open-tube, fluidized-bed mixing rate increased strongly with bed height; this effect is discussed further in Par. 7 below.

Figure 6
EFFECT OF SIZE OF SPHERICAL PACKING ON
SOLIDS MIXING IN A FLUIDIZED-PACKED BED

108-6189 Rev. 2

5. Effect of Cylindrical Packing

The solids diffusivities of 0.0038-in. fluidized particles in voids of $1/2 \times 1/2$ -, $3/8 \times 3/8$ -, and $1/4 \times 1/4$ -in. cylindrical packing are plotted in Figure 7. The slopes for these curves are somewhat less than for the spherical packing (Figure 6). Aside from the difference in shape, the larger cylinders exhibited a greater tendency for "bridging" when randomly packed. This bridging resulted in a nonuniform void structure throughout the bed and caused slightly erratic results.

Figure 7
MIXING DIFFUSION COEFFICIENTS FOR A FLUIDIZED-PACKED
BED WITH CYLINDRICAL PACKING

To determine whether length of time of fluidization affected the solids diffusivity, a run was made with the $1/4 \times 1/4$ -in. cylindrical packing in which samples were taken after 2.15, 3.84 and 4.84 min of fluidization. The diffusivity was found to be time-invariant to within about 1.5%.

6. Effect of Fluidized Particle Size

Runs were made with -40 +50, -60 +70, -100 +120, and -140 +170 mesh copper and nickel in the voids of $3/8$ -in. spherical packing to determine the effect of particle size on mixing rates. Diffusivities versus superficial gas velocities for the various particle sizes are plotted on Figure 8. The smaller fluidized particles showed the higher diffusivity.

Figure 8
EFFECT OF FLUIDIZED PARTICLE
SIZE ON SOLIDS MIXING

108-7084 Rev.

The previous runs were made with narrow cuts in screen sizes. Several runs were made with -120 +200 mesh particles, which have approximately the same average diameter as -140 +170 mesh particles (0.0038-in.) but have a greater spread in size distribution. Figure 9 compares the effect of particle distribution on the solids mixing for fixed packings of $1/2$ - and $1/4$ -in. spheres. Mixing is greater with

-120 +200 mesh particles, i.e., with the wider size spread. Also, the diffusion coefficient is no longer a simple logarithmic function of gas velocity. With the -120 +200 mesh particles, at increased velocities the fluidized bed segregated into two phases. There was a dilute, highly agitated phase in the upper portion of the packed bed, and a denser phase in the lower portion of the column.

Figure 9
EFFECT OF FLUIDIZED PARTICLE SIZE DISTRIBUTION ON SOLIDS MIXING

108-6188 Rev.

7. Effect of Bed Height for Fluidized and for Fluidized-packed Beds.

A brief examination was made of fluidized bed mixing, i.e., without the presence of a fixed packing. Figure 6 compares the mixing rates of a fluidized bed (static height 0.7 in.) and of the fluidized-packed beds. The mixing rate is greater for the fluidized bed because of the absence of fixed packing baffling effects.

With the fluidized bed, slugging increased with depth of bed. The slugging was accompanied by vigorous particle motion and an increase in the quantity of particles erupting from the surface of the fluidized bed. Runs were therefore made with the 0.0038-in. particles at a fluidizing gas velocity of 0.5 ft/sec to determine the effect of bed height on solids mixing for both the fluidized bed and the packed-fluidized bed. At initial bed heights of 0.7, 1.4, and 2.1 in., the corresponding diffusivities were 5.2, 14.8, and 27.8 ft²/sec. Figure 10 shows the relation of the solids diffusivity to bed height for the fluidized bed. The expansion ratio for the fluidized bed at this gas velocity was 1.4.

Figure 10
EFFECT OF BED HEIGHT ON
RATE OF SOLIDS MIXING IN
A FLUIDIZED BED

108-6190

The mixing diffusion coefficients for three different, initial, fluidized, particle bed depths ($1\frac{1}{8}$, $2\frac{1}{4}$, and $3\frac{3}{8}$ in.) for a fluidized-packed bed with $3/8$ -in. spherical packing were measured. Within the experimental accuracy, there was no effect of height of fluidized bed. The packing effectively baffles the slugs in the fluidized bed, which results in a smooth, uniform fluidization. In practical cases, mixing in open-tube fluidization should be considerably greater than in fluidized-packed beds.

8. Theoretical Models for Solids Mixing

Certain theoretical models were examined with respect to their ability to describe the mechanism of mixing of particulate solids in quantitative terms consistent with the experimental results. The three different diffusion models were: a molecular-kinetic model, a model developed for fluidization by Trawinski, and a "random-walk" model.

a. Molecular-kinetic Model

From visual observation, particles in a fluidized bed appear to move randomly. Therefore, it was considered, following Reboux,⁽⁴⁰⁾ that models developed for molecular motion might be appropriate.

If it is assumed that the particles do move in an independent random motion, the rate of solids mixing can be predicted by analogy with the kinetic theory of gases. The number of collisions per unit time, n , is given by the equation:

$$n = \underbrace{\frac{u\Sigma}{4(V_b - b)}}_{\text{wall collisions}} + \underbrace{\frac{\sqrt{2} \pi \nu d^2 u}{V_b - b}}_{\text{interparticle collisions}} \quad (3)$$

Knowing the collision rate and the mean speed of the particles, one can predict the diffusivity, D :

$$\begin{aligned} D &= (1/3) \frac{u^2}{n} \\ &= (1/3) u \frac{(V_b - b)}{\Sigma/4 + \sqrt{2} \pi \nu d^2} \end{aligned} \quad (4)$$

The volume V_b is determined by the void fraction of the packing, which was taken as 0.43 for the spherical packing used. The number and volume of the fluidized particles are determined from the void fraction of the particles at rest (0.36 was assumed) and the bed expansion ratio, which is a function of gas velocity.

For the 1/2-in. spherical packing and -140 +170 mesh (average diameter 0.0038 in.) fluidizing material, the following fluid-bed expansion ratios were obtained experimentally as a function of superficial gas velocity:

Superficial Gas Velocity (ft/sec)	Expansion Ratio
0	1.00
0.3	1.16
0.5	1.39
0.7	1.54
1.0	1.85
1.4	2.62

With the above bed expansion and void fractions, solids diffusion on the basis of a molecular-kinetics model can be predicted for various mean particle speeds. Figure 11 shows the solids diffusion coefficient as a function of gas velocity for this model, as calculated for the case in which the mean particle speed is equal to one-half V_0/ϵ , the mean velocity of gas in the packed bed voids.

Figure 11

COMPARISON OF THEORY AND DATA FOR
DIFFUSION OF FLUIDIZED PARTICLES IN
VOIDS OF 1/2-IN. SPHERES

108-6186 Rev.

b. Trawinski Model

Trawinski⁽⁵¹⁾ presented an equation for solids diffusivity based on a model in which individual particles pass through holes or openings in the particle suspension of the fluidized bed. Trawinski's equation is

$$D = \frac{K_s u}{6(1 - \epsilon_0)} \left(\frac{\Delta L}{L_0} \right). \quad (5)$$

The Trawinski equation was applied to the fluidized-packed-bed case of 1/2-in.-diameter spheres and 0.0038-in.-diameter fluidized particles to compare it with the other models. Figure 11 shows the solids diffusion coefficient, D , as a function of gas velocity, D being calculated for the case in which the particle velocity equals one-half V_0/ϵ , the average velocity of the gas in the packing voids.

c. Random Walk Model

Photographic work by Massimilla and Westwater⁽³¹⁾ revealed that the particles do not move individually but move in agglomerates. Rowe⁽⁴³⁾ has shown that overall particle motion results from small portions of the particles being carried up in the wake of individual bubbles. Small aggregates of particles fluidized in the voids of a packed bed would be carried up in the wake of the bubble and deflected in a random fashion at each encounter with an element of fixed packing. Baron⁽⁴⁾ predicted eddy diffusivities for gas in packed beds on the basis of lateral deflections of the stream caused by the presence of the packing. The theory of lateral deflections or random-walk displacements caused by the packing is here applied to lateral diffusion of solids in a fluidized-packed bed in a rectangular column.

The mean distance, $\overline{\Delta X}$, that a particle coming into contact with a sphere is deflected in the x direction (the direction of diffusion) is found by integration to obtain the average distance between the circumference and the chord perpendicular to the x direction that bisects the maximum horizontal circular cross section of the packing. Figure 12 shows a conceptual top and side view for a fluidized particle deflected by a spherical packing element. The mean distance, $\overline{\Delta X}$, may be expressed as

Figure 12
FLUIDIZED PARTICLE
DEFLECTION BY
SPHERICAL PACKING

108-6557

$$\Delta \bar{X} = \frac{\int_0^{\pi} (D_p/2) \sin \alpha d\alpha}{\int_0^{\pi} d\alpha} = \frac{D_p}{\pi}. \quad (6)$$

Actual solids mixing occurs as a batch system. In the model here adopted, both rising and falling particles are taken to have the same average lateral deflections.

The time, θ , between contacts for fixed packing arrayed rhombohedrally is found by dividing the vertical distance traveled between contacts by the mean particle velocity. That is,

$$\theta = 1/2 \frac{(1 + \sqrt{2}/3) D_p}{u} = \frac{0.909 D_p}{u}. \quad (7)$$

The average distance of deflection per contact and the time between contacts is related to a diffusion coefficient by the random-walk (mean free path) deflection in Einstein's diffusion equations originally developed for Brownian motion:

$$\overline{\Delta X^2} = 2D\theta \quad (8)$$

In the fluidized-packed bed the solids diffusivity would be

$$\begin{aligned} D &= 1/2 \frac{u}{0.909 D_p} \left(\frac{D_p}{\pi} \right)^2 \\ &= 0.0558 D_p u. \end{aligned} \quad (9)$$

Figure 11 shows the mixing diffusion coefficients as a function of gas velocity, as calculated for the cases in which the average particle velocities are one-half and one-tenth the average gas velocity in the packing voids.

The random-walk model should apply over the entire range of gas fluidization velocities for lateral solids mixing. At low flow rates, when the bubbles are uniformly distributed with little interaction, the particles carried in the bubbles would be deflected by the packing. At high flow rates, even though there may be interaction and coalescence of bubbles, the solids movement would still be in streams of particle aggregates that undergo lateral deflections by the packing. The random-walk model only applies to lateral solids mixing and not to axial mixing.

d. Comparison of Theoretical Models

The comparable cases calculated for each of the three models considered are shown in Figure 11, together with a plot of the experimentally-measured diffusion coefficients. The models based on Trawinski's theory and on the kinetic theory differ from the experimental data by factors of approximately 10 and 10^2 , respectively. The random-walk model appears to give a much closer approximation, especially when it is considered the experimental value falls within the two assumed cases of mean particle velocity (viz., one-half and one-tenth the average gas velocity).

Equation 9 indicates that the mixing diffusion coefficients for fluidized particles of a given size would be directly proportional to the diameter of the fixed packing. The

Figure 13
DEPENDENCE OF SOLIDS DIFFUSIVITY
ON FIXED PACKING DIAMETER

108-6192

dependence of the diffusion coefficient, D , on the spherical packing size, D_p , is shown in Figure 13 for the -140 +170 mesh particles fluidizing in the voids of various sizes of spherical packing. In Figure 13, the ratio D/D_p is plotted against the gas velocity. Figure 13 indicates that a linear proportionality holds between diffusivity and packing size. This is consistent with the random-walk theory for the case of particle velocity being independent of packing size.

Figure 11 indicates that the random-walk model is best in predicting the order of magnitude of the diffusion coefficient, and further verification of the model is shown in Figure 13. However, the predicted values shown on Figure 11 are based on estimated mean particle velocities.

Experimental measurements of particle velocities and a theoretical predication of particle velocities will be described in Par. 9 below.

9. Measurement of Average Particle Velocity

The diffusion coefficient for solids mixing, D , is related to the average fluidized particle velocity by the random-walk theory by rearranging Equation 9 as follows:

$$u = \frac{D}{0.0558D_p} \quad (10)$$

From the above equation and the diffusivity data, an average fluidized particle velocity can be calculated. The particle velocity for the -40 +50, -100 +120, and -140 +170 mesh particles are shown in Figure 14. The particle velocity decreases as a function of fluidizing gas velocity with increased particle weight.

Figure 14
AVERAGE FLUIDIZED
PARTICLE VELOCITY

108-6183 Rev.

As a means of independent measurement of the particle velocity, high speed motion pictures were taken of the fluidized particles. Approximately one square inch area of the particle bed at the wall was photographed at 4,000 frames/sec. One roll of film was examined for each run condition. At a given instant it was observed that large areas of the bed were relatively stagnant except where there were local disturbances caused by the bubbles. No area was permanently inactive; all areas would eventually be disturbed by passing bubbles. It was estimated that at a given instant for the beds of smaller particles, at the higher velocities, that about 50% of the bed was not in motion. At low velocities, with the 0.0141-in. particles, about 90% of the bed was inactive. In determining the particle velocities, the speed of the particles in the upward and downward directions, and the fraction of particles not in motion, were averaged in the following manner:

$$u_{ave} = \left(\frac{ave\ u_{up} + ave\ u_{down}}{2} \right) (\text{active fraction}) \quad (11)$$

The movement of the particles appeared not as random movement of individual particles but as streams of particles moving in the upward direction or falling in the downward direction caused by displacement by bubbles rising through the packed bed. Only the vertical velocity component was measured.

The average particle velocity based on the photographic data is only approximate for three reasons: (1) only a small sample of particle motion in terms of time and area was examined, (2) the proportion of particles in motion at a given instant was only estimated, and (3) the particle motion at the wall may not have been entirely representative of the internal motion. However, as shown in Figure 14, the particle velocity indicated by the diffusion theory and that estimated by photographic means are in close agreement.

a. Theoretical Prediction of Particle Velocities

The random-walk theory described the mechanism of lateral solids mixing, and therefore, the mixing diffusivities could be related to the size of the fixed packing and the mean particle velocity.

While the particle velocities were measured for the systems studied here, a generalized correlation requires a prediction of mean particle velocity based on the size and density of the fluidized particle and the properties of the fluidizing gas. To reveal the effect of these properties on the solids mixing, a theory is now presented that relates the average particle velocity to the fluidizing gas velocity.

b. Determination of Average Particle Velocity

The analysis of Rowe⁽⁴³⁾ and co-workers has shown that the particles are transported by the drag forces in the wake of bubbles rising through the fluidized bed.

This concept was used as the basis of a model for particle movement. The model adopted here is defined by these assumptions:

1) Gas flow in excess of the minimum velocity required for fluidization forms bubbles; that is, the volumetric rate of bubble formation is $W - W_{mf}$.

2) The ratio of the volume of particles moved by the bubbles to the volume of the bubbles is a constant F' for a given particle-gas system; that is, the volumetric rate of particles in motion is equal to $F'(W - W_{mf})$ for particles of a particular size. Implicit in this model is the assumption that the ratio F' is independent of $W - W_{mf}$. This assumption is equivalent also to asserting that F' is independent of bubble size as well as frequency.

10. Fraction of Particles Moved by Bubble

From the above assumptions, the volumetric rate of particles per unit area dragged by gas flow through the bubble can be expressed as

$$F' \left[\frac{W - W_{mf}}{S\epsilon} \right] = F'(V - V_{mf}), \quad (12)$$

where

$$F' = \frac{\text{volume of particles moved}}{\text{volume of gas forming bubbles}}.$$

The volume, F' , of particles moved will depend upon the force exerted on the particles (drag force) and the weight of each particle. Figure 15 and Table I describe the ratio of drag force divided by particle weight, calculated as discussed below, for four different particle sizes. Rowe and Henwood⁽⁴⁵⁾ have shown that the drag coefficient for a particle in an array of surrounding particles is 68.5 times the drag coefficient for an isolated particle. On this basis, Rowe⁽⁴⁴⁾ has shown that the minimum fluidization velocity can be calculated in the same manner as the terminal falling velocity by correcting the drag coefficient for an isolated sphere by this factor of 68.5. Therefore, it would follow that the minimum fluidization velocities for the different particle sizes would be of the same ratio as the calculated drag ratios. Figure 15 and Table I give the ratio of the drag

Figure 15
DRAG ON FLUIDIZED PARTICLES

force on the particle divided by particle weight for each particle size as calculated for the various conditions of gas velocity. As can be seen from Figure 15, this drag ratio increases with gas velocity for each particle size, the smaller particles having the higher rate of increase. In Table I also, further comparison is made of the relative drag ratios (relative to the 0.0054-in. particles) for each value of gas velocity. That is, at every gas velocity, the relative drag ratio $(\text{drag})_d / (\text{drag})_{0.0054}$ was calculated. The 0.0054-in. particles and the other particles were compared for the minimum fluidization gas velocity. These values (shown in Table I) are close to those of the relative drag ratios, as would be expected from the theoretical analysis presented in the paragraph above.

Table I
DRAG RATIOS FOR DIFFERENT COPPER-NICKEL PARTICLE SIZES

Fluidizing gas: N₂ at 20°C
 $\rho = 0.0724 \text{ lb/ft}^3$
 $\mu = 0.0175 \text{ cp}$

Particle Size		Particle Weight, M_p , lb	Minimum Fluidization Velocity, V_{mf} ft/sec	Velocity, ft/sec	$dV\rho/\mu$	Drag, Pounds/Pound $68.5 \frac{C_D \rho V^2}{2g} \frac{\pi d^2}{4} \frac{1}{M_p}$	Relative Drag	Relative Minimum Fluidization Velocity, $V_{mf} 0.0054/V_{mf}$
Mesh	In.							
-140 +170	0.0038	0.0922×10^{-7}	0.122	0.276	0.541	2.46	1.97	2.24
				1	1.96	10.13	1.95	
				2	3.92	27.5	1.89	
				3	5.88	37.1	1.87	
				4	7.84	52.5	1.83	
				5	9.80	69.7	1.81	
-100 +120	0.0054	0.265×10^{-2}	0.276	0.276	0.768	1.25	1.00	1.00
				1	2.78	5.21	1.00	
				2	5.56	11.9	1.00	
				3	8.35	19.9	1.00	
				4	11.1	28.7	1.00	
				5	13.9	38.5	1.00	
-60 +70	0.00905	1.75×10^{-7}	0.706	0.276	1.29	0.462	0.364	0.391
				1	4.67	2.04	0.391	
				2	9.34	4.83	0.406	
				3	14.0	8.20	0.412	
				4	18.7	12.05	0.420	
				5	23.3	16.4	0.426	
-40 +50	0.0141	4.72×10^{-7}	1.56	0.276	2.01	0.201	0.161	0.177
				1	7.27	0.934	0.174	
				2	14.5	2.29	0.193	
				3	21.8	3.97	0.200	
				4	29.1	5.96	0.208	
				5	36.3	8.15	0.211	

Since the minimum fluidization velocities are experimentally determined for each particle size and there is some question as to whether an average sieve opening size gives the appropriate particle size, the ratios determined on the basis of minimum fluidization velocities will be assumed to be the proper basis for estimating the relative volume of material dragged in the bubble wake.

The proposed method of predicting the movement of solids for any particle size is based on the above considerations. The volumetric rate of particles moved by the dragging action of bubbles is given by the product of the excess gas velocity ($V - V_{mf}$) for the given sized particle, times the constant F determined for the reference particle system (-100 +120 mesh, 0.0054-in. mean diameter), times the factor $V_{mf} 0.0054/V_{mf}$ correcting for relative drag ratios. Therefore, using the -100 +120 mesh particles as a basis, the volume of particles dragged by the gas flow per unit area through the bubble, V_p , is

$$V_p = F \left[\frac{V_{mf} 0.0054}{V_{mf}} \right] (V - V_{mf}), \quad (13)$$

where F = volume of particles moved/volume of gas forming bubbles for 0.0054-in.-diameter reference particle system.

11. Absolute Bubble Velocity

The rate of fluidized solids movement depends not only on the proportion of particles moved, associated with the excess gas velocity, but also on the absolute velocity of the bubble rising through the bed. The bubbles in a fluidized bed do not contain the same element of gas as do bubbles rising through a liquid, but rather are voids or openings surrounded by porous walls of particles through which there is a continual flow of gas. For this reason, and because there are complex pressure gradients that alter the volumetric gas flow, the absolute bubble velocities cannot be determined directly from the bed expansion and excess gas flow. However, ratios of bubble velocities can be estimated from the ratios of bed expansions for the various particle sizes.

This estimation would assume a) that essentially all of the bed expansion is due to the bubbles, and b) that changes in gas volumes because of the pressure effects in the bubbles at the same $V - V_{mf}$ for different sized particles are essentially the same.

The fluidized bed void fraction or the bubble volume can be calculated from the expansion ratio:

$$\epsilon_B = \Delta L / (\Delta L + L_0) = (R - 1) / R. \quad (14)$$

Figure 16 shows that the different particle sizes have about the same expansion ratio as a function of $V - V_{mf}$, except for the smaller -140 +170 mesh particles. For the -140 +170 mesh particles, the expansion is greater for a given $V - V_{mf}$, and it is concluded that the bubble velocities are lower in the following manner:

$$\text{Volumetric bubble rate} = S(V - V_{mf}) P, \quad (15)$$

where P is a factor for effect of pressure;

$$\text{Bubble residence time} = \frac{\Delta L S}{S(V - V_{mf}) P}; \quad (16)$$

$$\begin{aligned} \text{Bubble velocity} &= (L + \Delta L) / \frac{\Delta L S}{S(V - V_{mf}) P} = \left[\frac{L + \Delta L}{\Delta L} \right] (V - V_{mf}) P \\ &= \frac{(V - V_{mf}) P}{\epsilon_B}. \end{aligned} \quad (17)$$

Figure 16
BED EXPANSION RATIOS

Using the -100 +120 mesh (0.0054-in.) particles as a basis of comparison, and from the above assumption that the P factor is essentially the same for different particle size at the same $V - V_{mf}$, the relative bubble velocity between different particle sizes is

$$\frac{V_B}{V_{B \ 0.0054}} = \frac{\epsilon_{B \ 0.0054}}{\epsilon_B} \quad (18)$$

Table II compares relative bubble velocities.

Table II
COMPARISON OF BUBBLE VELOCITIES

Particle Size, mesh	V, ft/sec	V - V _{mf} , ft/sec	R	ϵ_B	$\epsilon_{B \ 0.0054}$	$\frac{\epsilon_{B \ 0.0054}^*}{\epsilon_B}$
-140 +170	0.62	0.50	1.22	0.180	0.145	0.807
	1.14	1.02	1.43	0.301	0.231	0.767
	1.55	1.43	1.69	0.408	0.310	0.760
	2.23	2.11	2.16	0.537	0.459	0.855
	2.96	2.84	3.18	0.685	0.603	0.880
-100 +120	0.62	0.34	1.11	0.099	-	1.00
	1.14	0.86	1.24	0.193	-	1.00
	1.55	1.27	1.41	0.291	-	1.00
	2.23	1.95	1.73	0.422	-	1.00
	2.85	2.57	2.22	0.549	-	1.00

*Comparison made at same $V - V_{mf}$.

The average particle velocity, u , can be determined from the amount of particles carried in the bubble wake and the relative bubble velocity as follows:

$$u = [\text{Volume of particles moved/area-time}][\text{Relative bubble velocity}]$$

$$= F \left[\frac{V_{mf0.0054}}{V_{mf}} \right] (V - V_{mf}) \left[\frac{\epsilon_{B0.0054}}{\epsilon_B} \right]. \quad (19)$$

Equation 19 presents the average particle velocity in terms relative to the 0.0054-in.-diameter copper-nickel shot. Since $V_{mf0.0054}$ is constant, a correlation of D is suggested in the following manner by substitution into Equation 9:

$$D = K(0.0558 D_p) \left[\frac{V - V_{mf}}{V_{mf}} \right] \left[\frac{\epsilon_{B0.0054}}{\epsilon_B} \right], \quad (20)$$

where

$$K = (F) (V_{mf0.0054}).$$

The experimentally obtained diffusivities versus $0.0558 D_p (V - V_{mf}/V_{mf}) (\epsilon_{B0.0054}/\epsilon_B)$ are plotted on Figure 17. The points deviate up to

a factor of two from the solid line, but considering that a range of approximately two orders of magnitude is covered, the theory does quite well. For each particle size, the points appear to have about the same slope as the solid line, and therefore deviate by a constant factor. This simple theory neglects any effects that may be associated with the rheology of the fluidized particles, which could account for the deviation of each particle size. From Figure 17 the value for F is 0.22. Therefore, for the 0.0054-in.-diameter particles, the amount of particles dragged is approximately one-fifth the bubble volume.

If minimum fluidization velocity data for a particular fluidization system are not available, the method of Rowe⁽⁴⁴⁾ can be used for predicting minimum fluidization velocities. Rowe's method was applied to the materials described here, and minimum fluidization velocities were predicted fairly well (see Table III).

Table III

CALCULATION OF MINIMUM FLUIDIZATION VELOCITIES

Fluidized Particle Size		$(C_{DR_e^2})_{mf}$	Re_{mf}	Calculated V_{mf} , ft/sec	Observed V_{mf} , ft/sec
Mesh	In.				
-140 +170	0.0038	5.81	0.24	0.122	0.122
-100 +120	0.0054	16.58	0.64	0.230	0.276
-60 +70	0.00905	78.6	2.7	0.578	0.706
-40 +50	0.0141	297	7.8	1.072	1.56

However, prediction of bed expansion is more difficult. Davies and Robinson⁽¹³⁾ concluded that, at present, no generally applicable correlation of expansion of aggregatively fluidized beds exists. Bed expansion depends upon the bubble formation. Bubble size would be affected by the type of gas distributor, column configuration, and bed height, as well as the properties of the fluidized particles and fluidizing gas.

However, as can be seen from Table II, the bed expansion effect is not large and could be neglected without too much error. Figure 18 is a plot of the experimental diffusivities versus $0.0558D_p(V - V_{mf})/V_{mf}$. This gives essentially as good a correlation as that on Figure 13, but in simpler terms.

On the basis of these data and arguments, the following generalized correlation for particle diffusivities results:

$$D = 0.22(0.0558D_p) \left[\frac{V - V_{mf}}{V_{mf}} \right] (V_{mf}^{0.0054}) \left[\frac{\epsilon_B^{0.0054}}{\epsilon_B} \right]. \quad (21)$$

Figure 18

GENERAL CORRELATION OF SOLIDS MIXING
IN A FLUIDIZED-PACKED BED NEGLECTING
RELATIVE BED EXPANSIONS

108-7307 Rev.

This development of a generalized correlation permits prediction of solids mixing rates in fluidized-packed beds for systems involving a different fluidizing gas and a different fluidized material than the nitrogen and copper-nickel system in which mixing measurements were made here. This generalized correlation is further verified by the measurement of thermal diffusivities for several fluidized materials and different fluidizing gases in the section on lateral effective thermal conductivities (Sec. IV-A).

12. Conclusions

- 1) The lateral mixing behavior of particles fluidized in the voids of a packed bed is not analogous to the random motion of gas molecules but rather to eddy diffusion in a flowing gas stream.
- 2) A random-walk model may be used to relate these solids diffusivities to the size of the fixed packing. Mean particle velocities

calculated from this theoretical model appear consistent with photographic observations of particle velocity. Depending on the size of the particle, the particle velocity is a factor of four to 200 lower than the average linear gas velocity in the packed bed. The mean particle velocity was independent of the packing diameter.

3) A generalized correlation for lateral solids diffusivity in a fluidized-packed bed with spherical packing was developed and has the following form:

$$D = 0.22(0.0558 D_p) \left[\frac{V - V_{mf}}{V_{mf}} \right] (V_{mf}^{0.0054}) \left[\frac{\epsilon_B^{0.0054}}{\epsilon_B} \right].$$

4) The rate of change of solids diffusivity with gas velocity is less for cylindrically packed beds than for spherically packed beds.

5) The rate of solids mixing increases with bed height for open-tube fluidization (no packing), but is virtually independent of height for fluidized-packed beds within the limits of the experimental conditions.

B. Axial Solids Mixing

Stagewise reactor processes and other processes involving the addition and removal of fluidized particles depend upon the vertical movement of the fluidized particles. The characteristics of axial solids mixing are important to the two-zone oxidation-fluorination system for processing uranium dioxide fuel, where the uranosic oxide (U_3O_8) fines are produced by oxidation of the uranium dioxide and then carried up in the fluidized bed to the upper fluorination zone to be fluorinated to uranium hexafluoride.

Measurements of axial mixing in fluidized-packed beds were made in similar manner as the measurements for the lateral mixing. Copper and nickel shot were again used for the fluidizing material. The following parameters were investigated for their effect on the longitudinal diffusivity of particles fluidizing in the voids of fixed packing:

- 1) Gas velocity
- 2) Fluidized particle size
- 3) Bed height
- 4) Fixed packing size
- 5) Fixed packing array.

1. Apparatus and Procedure

A 2-in.-ID Lucite column 4.5 ft high was used as a fluidization vessel in these tests. Sample ports were installed at 4-in. intervals along the length of the column to allow withdrawal of the fluidized material through

hypodermic needles. Humidified nitrogen distributed by a sintered metal plate at the base of the column was the fluidizing gas. Copper and nickel shot of the following sizes were used as fluidized material: -40 +50 mesh, -100 +120 mesh, and -140 +170 mesh. The fixed packings were 1/4-, 3/8-, and 1/2-in.-diameter spheres.

Equal weights of copper and nickel were charged to the column: first a lower layer of pure copper, and then an upper layer of pure nickel was charged. Runs were also made with nickel and copper charged in the reverse order, and the results indicated no significance in the order of charging. By means of a quick opening toggle valve and a rotameter, the bed was fluidized over a measured time period at a desired flow rate.

For the initial runs, extent of mixing was determined by analyzing sample concentrations at various bed heights by magnetic separation. From the concentration profile, the longitudinal mixing diffusivities were determined from the boundary value solution of the diffusion equation presented by Equation 2 in Sec. III-A-1:

$$C = 1/2 + \frac{2}{\pi} \left\{ \exp \left[-\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{H^2} \right] \sin\left(\frac{\pi x}{2H}\right) + (1/3) \exp \left[-9\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{H^2} \right] \sin\left(\frac{3\pi x}{2H}\right) + (1/5) \exp \left[-25\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{H^2} \right] \sin\left(\frac{5\pi x}{2H}\right) + \dots \right\}. \quad (2)$$

The experimental concentration profiles were found to be in good agreement with the profiles expected from this diffusion model (see Figure 19). After it was verified by the sample concentration profiles that diffusivities could be used to characterize the mixing rates, a simpler experimental procedure was used for the subsequent runs. In these later runs, the upper and lower halves of the bed were withdrawn separately, and each half was analyzed for overall (average) concentration. The weight fraction of nickel in the upper half was then averaged with the weight fraction of copper in the lower half. The solids diffusivity was then calculated from the following equation:⁽⁴⁶⁾

$$\bar{C} = 1/2 + \frac{4}{\pi^2} \left\{ \exp \left[-\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{H^2} \right] + (1/9) \exp \left[-9\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{H^2} \right] + (1/25) \exp \left[-25\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{H^2} \right] + \dots \right\}, \quad (22)$$

where $\bar{C}$ is the average concentration of copper in the lower half of the bed (or nickel in the upper half). Equations 2 and 22 are so related geometrically that D , θ , and H are identical. (H is half the height of the expanded fluidized bed.)

Figure 19

CONCENTRATION PROFILE FOR LONGITUDINAL SOLIDS MIXING IN A FLUIDIZED-PACKED BED

Fixed packing: 3/8-in.-diam steel spheres
 Fluidized material: -100 +120 mesh copper-nickel mixture (2800 g)
 Fluidized bed height: 24 in. (static)
 ~42 $\frac{3}{4}$ in. (expanded)
 Fluidizing gas: nitrogen (humidified)
 Superficial gas velocity, V_0 : 1.06 ft/sec
 Gas velocity (corrected for packing void fraction, $\epsilon = 0.442$), V_0/ϵ : 2.40 ft/sec
 Run duration: 1.75 min

Figure 19 gives the results of analysis of an experiment in which both of the above methods were used; that is, a concentration profile was measured, and the overall concentration of withdrawn halves of the fluidized bed was determined. Using the diffusivity determined from Equation 22, the concentration profile was calculated by means of Equation 2 and is shown by the solid line in Figure 19. The agreement of the line with the experimental points indicated that the simplified method for determining the diffusivities was accurate and that the diffusion equations were adequate for determining the rates of solids mixing. Also shown on Figure 19 are dashed lines indicating the effect of 10% deviation in D on the theoretical concentration profile.

2. Theory for Longitudinal Solids Mixing

A theoretical model can be developed for longitudinal solids mixing based on the following assumptions used for the model for lateral solids mixing:

- 1) Gas flow in excess of the minimum velocity required for fluidization forms bubbles. On a unit cross-sectional area basis, the volumetric rate of bubble formation is $V - V_{mf}$.
- 2) The ratio of the volume of particles moved by the bubbles to the volume of bubbles is a constant F' for a given particle-gas system; that is, the rate of particles in motion is equal to $F'(V - V_{mf})$.
- 3) The frequency of bubble formation is fairly constant with changing gas flow, in the range of interest. Therefore, increasing the gas flow, $V - V_{mf}$, causes a corresponding increase in average individual bubble volume.

From the theoretical development for lateral movement in fluidized-packed beds, it was determined that the volume of fluidized particles dragged by the gas flow through the bubble per unit area is

$$K(V - V_{mf})/V_{mf}, \quad (23)$$

where $K = F' V_{mf}^{0.0054}$.

For 0.0054-in.-diameter copper-nickel particles, the amount of particles dragged is approximately one-fifth the bubble volume. Darwin⁽¹²⁾ determined that the drift volume for particles of an ideal fluid disturbed by the movement of a spherical body is half the volume of the sphere. (The drift volume is the amount of fluid elements displaced by the passage of a sphere through the fluid.) Lighthill⁽²⁹⁾ further developed Darwin's considerations of drift of the elements of an ideal fluid caused by the movement of a sphere. The total drift of the elements of an ideal fluid is approximately proportional to the square of the radius of the sphere. Therefore, it is estimated that the total vertical drift for fluidized particles would be expressed as

$$\Delta X = K' \left(\frac{V - V_{mf}}{V_{mf}} \right)^{2/3}. \quad (24)$$

Einstein's equation relating diffusivity to mean free path and particle velocity is

$$D = (1/2) \bar{\Delta X} u. \quad (25)$$

Combining Equations 23 and 24 enables the vertical mixing diffusivity for fluidized beds to be expressed as

$$D = K' \left(\frac{V - V_{mf}}{V_{mf}} \right)^{5/3} \quad (26)$$

From this model, the following effects of the parameters would be predicted:

a. Gas Velocity

Figure 20 depicts the general form that diffusivity would have as a function of gas velocity. It would be expected that there would be no solids movement until the gas velocity attained the minimum fluidization velocity, and then the diffusivity would increase proportionally to $[(V - V_{mf})/V_{mf}]^{5/3}$ until the bubble size was either limited by the walls of the column or by its maximum stable size. Where the bubbles attain their maximum size, the diffusivity curve would be expected to level off with increasing velocity.

Figure 20
EFFECT OF GAS VELOCITY
ON LONGITUDINAL SOLIDS
MIXING BASED ON
THEORETICAL MODEL

b. Fluidized Particle Size

Harrison, Davidson, and de Kock⁽²¹⁾ have shown that the maximum stable bubble size is essentially proportional to the diameter of the fluidized particle. The bubble size and, hence, the vertical drift would therefore be expected to tend to increase with particle size. However, the weight of the particle, and therefore the resistance to movement, increases with particle diameter. For fluidizing materials of identical densities, longitudinal diffusivity could increase and attain a maximum, and then decrease with increasing fluidizing particle diameter.

c. Bed Height

Botterill and Bloore⁽⁸⁾ reported that average bubble size increases with bed height because of coalescence of smaller bubbles. Diffusivity should then increase with bed height, because of increased average bubble size and resultant increased drift, until the bubbles attain their maximum stable size.

d. Fixed Packing Size

Although the mean free path of lateral movements of the fluidized particles is determined by the diameter of the fixed packing, the mean free path of vertical movement would not necessarily be dependent on packing size. The vertical mean free path would be a function of the drift caused by the rising bubble. The axial diffusivities should be fairly independent of the fixed packing diameter. However, smaller packing would have a greater effect in dispersing the bubbles, and the axial diffusivities would also be affected to the degree that the bubble size is affected by the size of fixed packing.

e. Fixed Packing Array

Loosely packed (large voids) spheres would not disperse the bubbles as much as a tightly packed bed. Therefore, the diffusivities for loosely packed beds of fixed packing would be somewhat greater than for those of tightly packed beds. For randomly packed beds, there would be some differences in diffusivities with different charges because of variations in packing densities.

3. Experimental Results

a. Effect of Gas Velocity

Figures 21, 22, and 23 show axial mixing diffusivities as a function of gas velocity for three sizes of copper-nickel shot (-140 +170, -100 +120, and -40 +50 mesh, respectively) fluidizing in the voids of 3/8-in. spherical packing. For all runs, the fixed packing height exceeded the height of the fluidized particles in the expanded fluidized bed. The fluidizing gas velocities range up to a velocity of 10 ft/sec.

The shape of the initial portion of each curve and the break as a function of gas velocity is as shown in Figure 20. However, after the break in the initial portion the diffusivities tend to decrease and then rise with further increases in fluidizing gas velocity. This behavior after the first break in the curves could be attributed to confinement of large bubbles by the column walls (slugging). Since a 2-in. column was used, effects caused by the wall should be significant.

Figure 21

EFFECT OF BED HEIGHT ON LONGITUDINAL SOLIDS MIXING IN A
FLUIDIZED-PACKED BED OF -140 +170 COPPER-NICKEL SHOT

(-140 +170 Mesh Copper-Nickel Shot Fluidized in Voids
of 3/8-in. Spherical Packing)

Fluidizing gas: Nitrogen (humidified)

$$\epsilon = 0.442$$

Figure 22

EFFECT OF BED HEIGHT ON LONGITUDINAL SOLIDS MIXING IN A
FLUIDIZED-PACKED BED OF -100 +120 COPPER-NICKEL SHOT

(-100 +120 Mesh Copper-Nickel Shot Fluidized in Voids
of 3/8-in. Spherical Packing)

Fluidizing gas: Nitrogen (humidified)

$$\epsilon = 0.442$$

Figure 23

EFFECT OF BED HEIGHT ON LONGITUDINAL
SOLIDS MIXING IN A FLUIDIZED-PACKED BED
OF -40 +50 COPPER-NICKEL SHOT

(-40 +50 Mesh Copper-Nickel Shot Fluidized in
Voids of 3/8-in. Spherical Packing)
Fluidizing gas: Nitrogen (humidified)
 $\epsilon = 0.442$

Incipient fluidization is indicated at the beginning of each diffusivity-gas velocity curve as zero diffusivity.

b. Effect of Fluidized Particle Size

Higher solids diffusivities were obtained with -100 +120 mesh particles than for the -140 +170 and the -40 +50 mesh particles. This could possibly be because of the counteraction between the tendency for increased bubble size and hence increased drift with particle size, and the increased force to move larger particles as proposed earlier.

c. Effect of Bed Height

The static heights for the runs shown on Figures 21, 22, and 23 are 8, 16, and 24 in. As was expected, because of increased bubble coalescence and resultant increased average bubble size with increased height, the rate of increase of diffusivity with gas velocity in the initial portion of each curve increases with bed height.

Incipient fluidization is indicated at the beginning of each diffusivity-gas velocity curve as zero diffusivity.

d. Effect of Fixed Packing Size

Figures 24 and 25 show the effect of fixed packing size on the longitudinal diffusivity of -100 +120 and -140 +170 mesh particles. The static bed heights for the -100 +120 mesh and -140 +170 mesh particles were 16 and 8 in., respectively. The -100 +120 mesh particles were fluidized in the voids of 1/4-, 3/8-, and 1/2-in. spherical packing, and the -140 +170 mesh particles were fluidized in the voids of 3/8- and 1/2-in. spherical packing. For both fluidized particles sizes, there is an increase in diffusivity with increase in packing size. The considerations based on the theoretical model for longitudinal mixing indicate that the increased packing size has a reduced effectiveness in dispersing the rising bubbles. Hence, the drift of the particles would be greater.

Figure 24

EFFECT OF SIZE OF FIXED PACKING ON LONGITUDINAL
SOLIDS MIXING OF -140 +170 MESH PARTICLES AT
8-in. BED HEIGHT

Fluidized particles: -140 +170 mesh copper-nickel shot
Bed height (static): 8 in.
Fluidizing gas: Nitrogen (humidified)
Void fraction, ϵ : 0.442 for 3/8-in.-diam spherical packing
0.451 for 1/2-in.-diam spherical packing

Figure 25

EFFECT OF FIXED PACKING SIZE ON LONGITUDINAL SOLIDS MIXING OF -100 +120 MESH PARTICLES AT 16-in. BED HEIGHT

Fluidized particles: -100 +120 mesh copper-nickel shot
 Bed height (static): 16 in.
 Fluidizing gas: Nitrogen (humidified)
 Void fraction, ϵ : 0.428 for 1/4-in.-diam spherical packing
 0.442 for 3/8-in.-diam spherical packing
 0.451 for 1/2-in.-diam spherical packing

Figure 26 further reveals the baffling effect of the fixed packing. The -100 +120 mesh copper-nickel particles at an initial (static) bed height of 8 in. were fluidized in the voids of 3/8- and 1/2-in. spherical packing and in an open tube (no packing). The diffusivities were about an order of magnitude greater for the open-tube case than for the fluidized-packed-bed case. For fluidizing-gas velocities greater than about 2.3 ft/sec, the mixing was so rapid during a short time interval (< 0.5 min), that diffusion coefficients could not be determined by using the present method. Figure 26 indicates that the bubble dispersion or baffling by the fixed packing is quite significant in comparison with open-tube fluidization.

Figure 26

EFFECT OF FIXED PACKING SIZE ON LONGI-
TUDINAL SOLIDS MIXING OF -100 +120 MESH
PARTICLES AT 8-in. BED HEIGHT

Fluidized particles: -100 +120 mesh copper-
nickel shot

Bed height (static): 8 in.

Fluidizing gas: Nitrogen (humidified)

Void fraction, ϵ : 1.00 for no packing
0.442 for 3/8-in.-diam
spherical packing
0.451 for 1/2-in.-diam
spherical packing

e. Effect of Fixed Packing Array

For each of the above series of runs with the same fixed packing and fluidized particle size, the fluidized particles were drained and recharged without disturbing the packing; with this procedure, no information is obtained on variability of the packing structure. Four runs were made to determine the effect of variability of the void structure of the fixed-bed packing. Two of these runs were made at fluidized particle heights of 8 and 16 in. (static), with the same undisturbed fixed packing. Two other runs were made at the same two heights, but with separate charges of fixed packing. In each case, the fixed packing was randomly charged. The results of these tests are shown in Figure 27. There was as much as a factor of 1.8 difference in diffusivities between comparable beds, apparently because of the random variations in packing void structure.

Figure 27

EFFECT OF CHANGE IN VOID STRUCTURE OF FIXED PACKING ON LONGITUDINAL SOLIDS MIXING IN A FLUIDIZED-PACKED BED

Fluidized particles: -100 +120 mesh copper-nickel shot

Fixed packing: 3/8-in.-diam steel spheres

Fluidizing gas: Nitrogen (humidified)

f. Application to Reactor Design

The following generalized conclusions would have significance in reactor design:

- 1) Higher rates of mixing are attained with -100 +120 mesh particles than with larger or smaller particles.
- 2) Mixing increases with bed height.
- 3) Longitudinal mixing in fluidized-packed beds is much less than for open-tube fluidization.
- 4) Mixing is only slightly faster for 1/2-in. spherical packing than for 3/8-in. packing.
- 5) Random arrangements of the fixed packing have some effect on mixing rates. As a first approximation, these effects can be neglected. However, for a careful design analysis, a variability of a factor of two should be considered.

The effect of column diameter is not known. The discussion on the theoretical model would indicate that wall effects are quite significant. Also, work is needed to determine the effect of fluidized particle size on the

bubble size in order to obtain a generalized correlation for longitudinal mixing. A generalized correlation was developed for lateral-solids mixing because the lateral-solids-mixing model is independent of bubble size. However, the longitudinal drift is dependent upon bubble size.

C. Vertical Transport of Fines

Some cursory investigations were made on the vertical transport of fines in a fluidized-packed bed and in a fluidized bed. In the fluid-bed fluoride volatility processes, fines are produced by partial fluorination of the uranium dioxide pellets to form uranyl fluoride fines, and by oxidation of the uranium dioxide to form uranous oxide fines. The formation of fines is common to most fluidized-bed operations, either by chemical reaction or by attrition. Runs were made to estimate the rates at which fine particles are transported in a vertical direction from the bottom to the top of the column.

1. Apparatus and Procedure

Runs were made in the 2-in.-ID Lucite column used for the axial mixing studies and in a 3-in.-diameter brass column. A 1-in. strip along the length of the brass column wall was cut out and then sealed with Lucite to give a window for visual observation. The brass column was used to minimize any effect of static charges. The column was 6 ft long, with sample ports every 4 in. along the length. An auxiliary filter chamber was connected in the off-gas line from each column to collect elutriated fines.

Humidified nitrogen was the fluidizing gas. Because of the convenience of magnetic separation, nickel was used as the fine material. A Coulter Counter* analysis of the size distribution of the nickel fine particles is shown in Figure 28. On a weight-percent basis, the average diameter of

Figure 28
NICKEL FINES PARTICLE SIZE
ANALYSIS BY COULTER COUNTER

*Coulter Electronic Sales, Chicago, Illinois.

the nickel fines is 24 microns. Copper shot (-100 +120 mesh) and fused alumina grain (-40 +170 mesh) were used as the fluidized particles, and for the runs involving a fluidized-packed bed, 3/8-in.-diameter steel spheres were used as the fixed packing.

Because the anticipated rates of fines transport would be much greater than the mixing rates for the fluidized particles, the time intervals for sampling after initial fluidization would be very short. To improve precision by increasing the time of measurement, and to minimize elutriation losses, the total distance of diffusion was increased. In the previous studies, equal amounts of pure copper and nickel were charged to the fluidization column. For the runs in which fines transport was measured, a charge equivalent to a static bed height of 4 in. containing fines and fluidized particles was added to the bottom of the column, and then 8 to 32 in. of pure fluidized particles (without fines) was subsequently charged. It was then assumed that the fines diffused into an infinite medium. This results in a concentration profile following this equation:⁽¹¹⁾

$$\frac{C}{C_0} = \operatorname{erf}\left[\frac{a-x}{2\sqrt{D\theta}}\right] + \operatorname{erf}\left[\frac{a+x}{2\sqrt{D\theta}}\right], \quad (27)$$

where

C = Concentration of nickel at time θ ;

C_0 = Initial concentration of nickel;

θ = Time, sec;

x = Distance, ft;

a = Initial column height containing nickel, ft;

and

erf = Error function, defined as

$$\operatorname{erf}(z) = \frac{2}{\sqrt{\pi}} \int_0^z e^{-z^2} dz.$$

2. Two-in.-diameter Column

All the runs made in the 2-in. Lucite column were studies of nickel fines (mean diameter, 24 microns) transport in a fluidized-packed bed consisting of 3/8-in. spherical packing and of copper shot (-100 +120 mesh). The column was charged first with approximately 30 g of nickel fines mixed with enough copper shot (480 g) to be equivalent to 4 in. of static bed height. Then, an 8-in. (static) bed equivalent of copper was charged to the column above the nickel-copper mixture.

After the bed had been fluidized for 6 sec, the three 4-in. sections of the bed were withdrawn and analyzed for nickel concentration. The initial nickel concentration, C_0 , was based on the total amount of nickel recovered in the separation step, rather than on the amount charged. The nickel was recovered from the bed and from the auxiliary filter chamber. Approximately 85% of the nickel was accounted for after each run. The measured concentration of fines in each section was used to determine a concentration profile fit by Equation 27 with a suitable value for D . By this procedure, the fines-transport diffusion coefficients were estimated. Figure 29 compares the fines-transport diffusion coefficients with the mixing diffusion coefficients of the coarser fluidized particle medium. The transport diffusivities for the nickel fines (~ 24 microns) is one to two orders of magnitude greater than that of the copper short (~ 140 microns). These fines-transport diffusivities apply only to fines movement from the bottom to the upper sections of the bed.

Figure 29

COMPARISON OF VERTICAL
FINES TRANSPORT RATE WITH
LONGITUDINAL FLUIDIZED
PARTICLE MIXING RATES

Fixed packing: 3/8-in.-diam
spheres

3. Three-in.-diameter Brass Column

Alumina grain ($-40 +170$ mesh) was used as the fluidized material for the fines transport measurements with the 3-in.-diameter column. The runs were made with the 3-in. column. One run was made

with a fluidized-packed bed with 3/8-in., fixed, spherical packing. One hundred grams of nickel fines were thoroughly mixed with alumina grain (equivalent to a static 4-in. bed height) and charged to the bottom of the column. Pure alumina grain was then added after the initial nickel-alumina charge to provide a total bed height of 36 in. The fluidizing gas velocity was 0.804 ft/sec (corrected for packing voidage). Samples were withdrawn and analyzed for fines concentration after various time intervals of fluidization.

Figures 30 and 31 describe the concentration profile of the bed at various times. After about 30 sec, virtually complete mixing had occurred, and the equilibrium (final) concentration profile was attained.

Figure 30

CONCENTRATION OF FINES IN ALUMINA
GRAIN BED AFTER VARIOUS TIME
INTERVALS OF FLUIDIZED-PACKED-BED
OPERATION

Column: 3-in. diam
Fixed packing: 3/8-in.-diam steel spheres
Fluidized particles: -40 +170 mesh alumina grain, -500 mesh nickel fines (mean size, 24 microns) (initial concentration of nickel fines in nickel-alumina mixture in lower 4 in. of bed, 24.3 w/o)
Fluidizing gas: nitrogen (humidified)
Gas velocity (corrected for fixed packing voidage): 0.8 ft/sec

108-7272

Figure 31

EQUILIBRIUM CONCENTRATION PROFILE
OF FINES IN ALUMINA GRAIN RESULTING
FROM FLUIDIZED-PACKED-BED
OPERATION

Column: 3-in. diam
Fixed packing: 3/8-in.-diam steel spheres
Fluidizing particles: -40 +170 mesh alumina grain, -500 mesh nickel fines (mean size, 24 microns) (initial concentration of nickel fines in nickel and alumina mixture in lower 4 in. of bed, 24.3 w/o)
Fluidizing gas: nitrogen (humidified)
Gas velocity (corrected for fixed packing voidage): 0.8 ft/sec

108-7273

The fines-transport diffusion coefficient in the fluidized alumina was estimated from Equation 27 to be $8 \times 10^{-3} \text{ ft}^2/\text{sec}$ after 10 sec, and from the concentration profile at 20 sec, the fines diffusivity was estimated to be $14 \times 10^{-3} \text{ ft}^2/\text{sec}$. The differences between the diffusivities at 10 and 20 sec could be attributed to the fines movement not following a diffusion model. The fines were driven up the column, not only by a concentration differential, but also by the drag force associated with the gas flow. Therefore, there would be some time effect on the diffusivities if the fine particle movement were defined by a diffusion model that assumes equal diffusivities in either direction. Also, there could have been radial concentration variations, which were neglected, since samples were withdrawn only at the axis of the column.

The equilibrium concentration profile for the completely mixed fines is shown in Figure 31. This is similar to the fines concentration profile in fluidized beds reported by Thomas, Gray, and Watkins.⁽⁵⁰⁾ The fines concentration is higher toward the bottom and the top of the bed than at the center. At the bottom of the column, where bubbles are initially forming, fluidization is not as vigorous as it is higher in the bed. Less vigorous fluidization may be expected to be associated with a higher fines retention among the coarser fluidized particles. Because of the lighter weight of the fine particles compared to the larger alumina particles, the fines tend to be carried by the gas to the upper portion of the bed; this effect may account for the higher fines concentration at the top of the bed.

There was a large amount of scatter in the data. Since these tests were preliminary and few were made, the reason for this scatter has not been entirely determined. Adhesion between the fine particles and adhesion of the fine particles to the large particles might cause nonuniformities in fines distribution.

A run was also made in a fluidized bed (without packing) at a gas velocity of $0.333 \text{ ft}/\text{sec}$ and an initial (static) bed height of 26 in. The fines diffusivity for this case was estimated to be $2.5 \times 10^{-3} \text{ ft}^2/\text{sec}$.

4. Conclusions

The data on rates of fines movement in fluidized-packed and fluidized beds are rather preliminary but reveal that the fines transport from the bottom to the top of the bed is about an order of magnitude greater than the fluidized particle movement. Runs were made for just a few conditions. The rate of fines transport in the downward direction, rather than in the upward direction, is not known. The extent of the baffling effect exerted by the fixed packing on the fines movement is not known. Also, the effect of the size of the fine particles and the size of the coarser fluidized particles on the fines movement would need to be determined before a general correlation could be developed.

IV. HEAT TRANSFER

The fluidized-packed-bed concept was developed essentially because it would provide a system wherein large nonfluidizable particles undergoing an exothermic reaction would experience rapid heat removal. It was conceived that inert particles fluidizing in the voids of uranium dioxide pellets undergoing either fluorination or oxidation would increase heat transfer and hence improve reaction control. However, little basic heat-transfer data for fluidized-packed beds have been reported in the literature.

Sutherland et al.⁽⁴⁸⁾ were interested in fluidized-packed beds because the baffling effect of the fixed packing permits smooth fluidization (without slugging) for large height-to-diameter ratios. Preliminary investigations on the effect of fixed packing in fluidized beds, on the wall heat transfer coefficients were made by Sutherland, et al.

Ziegler and Brazelton⁽⁶⁰⁾ made a systematic study of heat transfer in fluidized-packed beds for various combinations of fixed packing and fluidized particle sizes. However, the fluidizing gas velocities that were used were generally below the range that would be used in processing.

Further studies were therefore made of heat transfer in fluidized-packed beds. A systematic study of lateral bed thermal conductivity was made that permitted the formulation of a generalized correlation. Some measurements were also made of longitudinal thermal conductivities, wall heat-transfer coefficients, and fixed packing surface heat-transfer coefficients in fluidized-packed beds.

A. Lateral Effective Bed Thermal Conductivity

For chemical reactors consisting of packed beds, temperature control depends upon the effective thermal conductivity of the packed bed. In packed beds, the major contributor to heat transfer is eddy mixing of the gas stream. In fluidized-packed beds, the major contributor to heat transfer is the mixing of the fluidized particles. To obtain a further understanding of the mechanism of fluidized particle heat transfer, lateral effective thermal conductivity of fluidized-packed beds was measured in a rectangular column, of the same dimensions as was used in the lateral solids mixing studies. This enabled a direct comparison of thermal diffusivities with the solids mixing diffusivities, and enabled the development of a generalized correlation of bed thermal conductivities for a fluidized-packed bed. The effects of fixed packing size, fluidized particle size and density, and different gases on lateral thermal conductivities, in a range of gas velocities that would be used in pilot plant processing, were investigated.

1. Apparatus and Procedure

The rectangular column used in this study (shown in Figure 32) was $1\frac{11}{16}$ in. wide, $8\frac{1}{2}$ in. deep, and 7 in. long. The two ends were aluminum plates to which copper cooling coils were bonded with a plastic heat transfer cement. The two long sides of the column were $1/2$ -in. thick transparent plastic Plexiglas, to which the cooling plates were bolted and cemented. The gas distributor was a $1\frac{3}{4} \times 6\frac{3}{4}$ -in. sintered stainless-steel plate, with neoprene gaskets sealing the edges.

Figure 32

SCHEMATIC DIAGRAM OF APPARATUS FOR MEASURING LATERAL THERMAL CONDUCTIVITIES IN A FLUIDIZED- PACKED BED

108-7643

Water was supplied to the cooling coils by a constant head tank. Water inlet and outlet temperatures were monitored by iron-constantan thermocouples. The water flow through the cooling coils was measured by collecting the exit water in a graduated cylinder during a known time interval. Air and helium were used as fluidizing gases. The fluidizing gas flow was measured by a rotameter having a range of 0 to 6.75 cfm for air at 70°F and 14.7 psia. The rotameter readings were corrected for helium by ratioing the square roots of the molecular weights of air and helium. For all runs, the Reynolds number for helium and air flowing past the float was in excess of 1000. This was well into the range of constant coefficient of discharge.⁽¹⁰⁾

The heater was an electrical resistance bayonet heater, 6 in. long, $1\frac{1}{2}$ in. wide, and $1/8$ in. thick. Current was supplied and regulated by a variable autotransformer. Power input was measured by a voltmeter and an ammeter. An iron-constantan thermocouple was spot-welded to one side of the heater to measure heater skin temperatures.

Bed-temperature probes were made by drilling holes into steel balls that made up the packing, and soldering the thermocouple ends inside the holes. The probes were placed at 3- and 6-in. heights in the bed. Three or four probes were placed at heights of 3 or 6 in. on each side of the heater, which was located in the center of the column. Because of the larger thermal conductivity in the vertical direction, compared with the horizontal direction, the temperatures were essentially uniform in vertical cross section.

The packed-bed height for all runs was 6 in. Packing consisted of $1/2$ -, $3/8$ -, $1/4$ -, and $1/8$ -in. steel spheres. For each run, the amount of fluidized particles was adjusted so that the height of the expanded fluidized bed was 6 in. (level with the top of the packing). If the fluidized portion of the bed were less than 6 in., the heat-transfer rates would have been somewhat lower because the bed of fluidized particles would not have extended throughout the entire height of the packing; and if the fluidized particles extended above the packing, the heat-transfer rates would have been somewhat higher because of the freer movement of the particles above the restrictions of the packing.

One series of runs was made with helium as a fluidizing gas. All other runs used air. Minus 40 +50 mesh, -60 +70 mesh, -100 +120 mesh, and -140 +170 mesh copper-nickel shot, -60 +70 mesh alumina, and -40 +50 mesh glass beads were used for fluidized material.

For the constant cross-sectional area, the Fourier equation relating rate of heat transfer to thermal conductivity can be integrated to

$$q = k A_s \frac{\Delta t}{\Delta x}, \quad (28)$$

where

q = Rate of heat transfer, Btu/(hr)(ft²);

k = Thermal conductivity, Btu/(hr)(ft²)(°F/ft);

A_s = Cross-sectional area, ft²;

and

$\Delta t/\Delta x$ = Temperature gradient, °F/ft.

The lateral temperature gradients were determined from the location and temperature of the probes in the fluidized-packed bed. The lateral temperature profiles were generally linear, and therefore heat losses to the gas stream and to the ambient air through the column walls could be assumed small. The effective bed thermal conductivities were based on the average of the temperature gradients on either side of the centrally located heater. The heat flux, q , was based upon the heat pick-up of the water in the cooling coils.

2. Experimental Results

Table IV lists the thermal conductivities and the thermal diffusivities for the different experimental conditions, as well as the physical properties of the fluidized particles, and the properties of the fluidized-packed beds. The thermal diffusivity is related to the thermal conductivity by

$$\alpha = \frac{k_e}{\rho_B C_p}, \quad (29)$$

where

α = Thermal diffusivity, ft^2/sec ;

k_e = Bed thermal conductivity, $\text{Btu}/(\text{sec})(\text{ft}^2)(^\circ\text{F}/\text{ft})$;

ρ_B = Fluidized bed density, lb/ft^3 ;

and

C_p = Fluidized particle heat capacity, $\text{Btu}/(\text{lb})(^\circ\text{F})$.

The fluidized-bed density is based on the total weight of fluidized particles divided by the volume of the bed, uncorrected for void fraction of the fixed packing. Since the heat is transported by the particles, the density must be based upon the number of particles per unit volume. The packing dilutes the number of particles, and hence the reduction of heat-carrying capacity must be compensated by including the volume occupied by the fixed packing.

The gas velocity is based on room temperature and pressure. The slight increase in velocity because of increase in gas temperature in the heated bed (average bed temperature about 50°C) was neglected.

One set of runs was made with $3/8$ -in.-diameter Lucite spheres to determine the importance of packing material on α . There was essentially no difference between the value of α for the Lucite-packed

Table IV
THERMAL CONDUCTIVITIES AND PROPERTIES OF THE FLUIDIZED-PACKED BEDS

ϵ taken as 0.415 for all packing								
			ρ , lb/ft ³	C_p , Btu/(lb) °F				
			Cu-Ni	555	0.105			
			Alumina	249	0.217			
			Glass	137	0.20			
Material	d, in.	D _p , in.	$\frac{V_0}{\epsilon}$, ft/sec	$\frac{k_e}{(hr)(ft^2)}$ °F/ft	ρ_B , lb/ft ³	$\alpha \times 10^4$, ft ² /sec	V _{mf} , ft/sec	Fluidizing Gas
Cu-Ni	0.0038	3/8	0.94	20.0	133	3.98	0.12	Air
			1.79	19.4	110	4.67		
			2.63	24.8	93.5	7.01		
			3.57	30.9	76.4	10.7		
Cu-Ni	0.0038	3/8	0.85	19.2	133	3.82	0.28	Helium
			2.09	24.0	110	5.78		
			3.65	24.4	93.5	6.90		
			4.84	28.6	76.4	9.90		
Cu-Ni	0.0054	1/2	1.52	16.7	126	3.52	0.28	Air
			2.63	42.0	111	10.1		
			3.86	65.9	92.4	18.9		
Cu-Ni	0.0054	3/8	0.94	6.5	125	1.38	0.28	Air
			1.52	11.0	113	2.58		
			2.07	14.2	101	3.72		
			2.63	22.8	96.4	6.25		
			3.23	32.0	86.2	9.84		
			3.86	39.2	77.6	13.4		
Cu-Ni	0.0054	1/4	0.94	5.3	117	1.20	0.28	Air
			1.52	6.2	105	1.56		
			2.07	11.5	94.5	3.22		
			2.63	14.1	84.0	4.44		
			3.23	20.4	76.0	7.12		
			3.86	19.1	68.5	7.37		
Cu-Ni	0.0054	1/8	1.52	3.6	85.1	1.12	0.28	Air
			2.63	5.6	65.4	2.28		
			3.86	21.4	44.7	6.25		
Cu-Ni	0.00905	3/8	1.52	3.1	114	0.70	0.71	Air
			2.07	4.7	106	1.18		
			2.63	7.7	98.5	2.07		
			3.23	9.8	93.4	2.75		
			3.86	12.4	84.1	3.90		
Cu-Ni	0.00905	1/4	1.52	2.2	113	0.52	0.71	Air
			2.07	2.6	105	0.65		
			2.63	3.5	97.1	0.94		
			3.23	5.0	87.0	1.52		
			3.86	5.5	81.0	1.78		
Cu-Ni	0.0141	3/8	1.90	1.3	129	0.26	1.56	Air
			2.11	1.4	127	0.30		
			2.44	2.0	123	0.44		
			2.59	2.4	121	0.53		
			2.96	2.5	117	0.57		
			3.17	5.5	114	1.28		
			3.53	4.5	112	1.08		
			3.86	10.2	109	2.47		
Cu-Ni	0.0141	1/4	2.07	1.0	131	0.21	1.56	Air
			2.63	1.5	123	0.32		
			3.23	1.8	114	0.42		
			3.86	1.9	109	0.46		
Alumina	0.00905	3/8	0.94	2.9	46.8	0.80	0.36	Air
			1.74	5.5	41.5	1.68		
			2.63	7.8	31.6	3.16		
Glass	0.0141	3/8	1.20	4.3	38.3	1.57	0.39	Air
			2.63	6.5	28.9	3.12		
			3.23	10.5	24.9	5.86		
			3.86	8.6	20.6	5.83		

bed and the steel-packed bed, although there is about a factor of 50 difference in the thermal conductivities of the two materials. This result is confirmed by Frischmuth,⁽¹⁶⁾ who found that packings of glass, steel, and copper spheres all yield the same heat-transfer properties.

3. Theory of Heat-transfer Mechanism

Since the heat is transported by the moving fluidized particles, the thermal diffusivity and the solids-mixing diffusivity should be related. Figure 33 compares the thermal diffusivities, α , for the copper-nickel shot with the solids-mixing diffusivities, D . Since the lateral motion of the particles depend upon the amount of deflection by the fixed packing, the diffusivities were divided by the fixed packing diameter, D_p . Experimental points for the thermal diffusivities are shown. The dashed lines represent the solids-mixing diffusivities. For each different particle size, the α 's are less than the D 's by about a factor of 2. This can be accounted for by the fact that the fluidized particles do not come to an

Figure 33

COMPARISON OF SOLIDS MIXING DIFFUSIVITIES WITH THERMAL DIFFUSIVITIES FOR COPPER- NICKEL FLUIDIZED PARTICLES

equilibrium temperature immediately with their surroundings as they move from one point of the bed to another. For a spherical particle, the rate of exchange of its sensible heat to its surroundings is

$$\frac{1}{6}\pi d^3 \rho C_p \frac{dT}{d\theta} = -h\pi d^2(T - T_s). \quad (30)$$

The temperature of the particle can be related to the time, θ , by integration as follows:

$$\int_0^\theta d\theta = \int_{T_1}^{T_2} - \frac{d\rho C_p}{6h} \frac{dT}{T - T_s}; \quad (31)$$

$$\theta = \frac{d\rho C_p}{6h} \ln \frac{T_2 - T_s}{T_1 - T_s}, \quad (32)$$

where

T_1 = Initial particle temperature,

and

T_2 = Final particle temperature.

For example, a 0.0054-in. copper particle moving at an average velocity of 0.4 ft/sec in an air-fluidized bed would have a Reynolds number, $du\rho/\mu$, of about 1. For this value of Reynolds numbers, the Nusselt number hd/k_g for spheres is approximately 2. Using this value of Nusselt number, the heat-transfer coefficient, h , for the particle surface would be about 72 (Btu)/(hr)(ft²)(°F), based on a k_g of air at an average temperature of 50°C. By substituting the above data in Equation 32, it may be estimated that the time required for a particle to transfer 50% of its heat is

$$\theta = - \frac{(5.4 \times 10^{-3})(8.9 \times 62.4)(0.1)}{(6)(12)(72)} (\ln 0.5)(3600) = 0.145 \text{ sec.}$$

The above calculation assumes rapid, internal, particle-heat transfer and, hence, uniformity of particle temperature. If k/rh exceeds 6, the internal heat transfer is very rapid, and there is essentially no internal temperature gradient.⁽²⁰⁾ For a 0.0054-in.-diameter copper particle,

$$\frac{k}{rh} = \frac{220 \times 12}{0.0027 \times 72} = 1.4 \times 10^4.$$

The value of 1.4×10^4 is well in excess of 6, and the assumption of no internal temperature gradient is justified.

Therefore, although the rate of particle movement is the same for both the heat and mixing studies, and since the transfer of sensible heat by the fluidized particles is not immediate, the rate of heat transfer is reduced. From the following series of equations it can be seen that the slow exchange of heat would reduce the thermal diffusivity in comparison to the mixing diffusivity. By definition,

$$q = k_e A_s \frac{dt}{dx}; \quad (33)$$

and

$$N = DA_s \frac{dC}{dx}; \quad (34)$$

where

N = Rate of mass transfer, lb/hr;

and

C = Concentration, lb/ft³.

Since the heat transfer is essentially effected by transport of the fluidized solids, the rate of heat transfer can be expressed in terms of the rate of solids mixing, as follows:

$$q = QN = -QDA_s \frac{dC}{dx}. \quad (35)$$

The term Q is the amount of heat content released by the fluidized particles as they move from different points in the fluidized-packed bed. Since the tracer for the particle concentration change in a system in which heat transport is being measured is temperature, the concentration gradient can be expressed in terms of a temperature gradient as follows:

$$q = \frac{(T_1 - T_2)}{(T_1 - T_s)} C_p \rho DA_s \frac{dt}{dx}, \quad (36)$$

where Q has been replaced by $[(T_1 - T_2)/(T_1 - T_s)] \rho C_p$, and T_s is the temperature of the surrounding bed. The temperature T_s is taken to be equivalent to t in the term dt/dx , and T_2 is the final temperature of the particle before moving to a new location. The term $(T_1 - T_2)/(T_1 - T_s)$ represents the fraction actually transferred of the total heat content that could thermodynamically be transferred by the particles as they change surroundings. It then follows that

$$k_e A_s \frac{dT_s}{dx} = \frac{(T_1 - T_2)}{(T_1 - T_s)} C_p \rho D A_s \frac{dT_s}{dx}; \quad (37)$$

and

$$\frac{(T_1 - T_2)}{(T_1 - T_s)} D = \frac{k_e}{\rho C_p} = \alpha. \quad (38)$$

If the particles were to reach equilibrium, the term $(T_1 - T_2)/(T_1 - T_s)$ would be 1. However, because the particles move from point to point before they attain the temperatures of the surroundings, $(T_1 - T_2)/(T_1 - T_s)$ is less than 1. Therefore, if the average residence time of a particle in one region is about 0.15 sec, α would be about one-half as large as D .

Figure 33 shows that at the lower gas velocities the thermal diffusivities approach the values of the solids-mixing diffusivities. At the lower gas velocities, the particle motion is less, a circumstance that results in a longer residence time for the particles to reach equilibrium.

The solids mixing data showed that for 0.0054-in. copper-nickel particles, the particle velocity increased for 0.06 to 1 ft/sec as the gas velocity increased from 0.6 to 4 ft/sec. If the bed were divided into $3/8 \times 3/8 \times 3/8$ -in. cells (based on $3/8$ -in.-diameter packing), the particle residence time in each cell would be in a range of 0.0311 to 0.52 sec. This is in line with the estimated residence time of 0.15 sec, which would result in α being about one-half of D .

Since the mechanism of heat transfer is the same as that of particle mixing, except for the added factor of slow heat release from particles, it would follow that the thermal diffusivities could be correlated in the same fashion as the mixing diffusivities. In Sec. III on solids mixing, a general correlation was developed for relating the solids mixing with the basic parameters of fixed packing size, minimum fluidization velocity, and fluidizing-gas velocity. From the properties of the fluidized particles and fluidizing gas, the minimum fluidization velocity can be predicted. Figure 34 correlates thermal diffusivity with $0.0558 D_p (V - V_{mf})/V_{mf}$. The correlation includes a range of spherical packing sizes from $1/8$ to $1/2$ in., fluidized-particle sizes from 0.0038 to 0.0141 in., fluidized particles of copper-nickel shot, alumina, and glass beads, and air and helium as fluidizing gases. Again, the slope of the curve is unity, giving support to the theoretical mechanism proposed in Sec. III.

The correlation can be expressed as

$$\frac{k_e}{\rho_B C_p} = \alpha = (0.04) 0.0558 D_p \left(\frac{V - V_{mf}}{V_{mf}} \right), \quad (39)$$

where all units are in feet and seconds.

Figure 34
GENERAL CORRELATION FOR THERMAL
DIFFUSIVITIES

108-7236 Rev.

However, of more value for engineering calculations is the thermal conductivity of the bed, rather than the thermal diffusivity. The thermal conductivity can be determined from the above correlation if the density of the fluidized bed is known. This density is a function of the gas velocity; that is, it involves bed expansion. At present, there is no good correlation for predicting bed expansion for gas-fluidized beds. Therefore, a modified thermal diffusivity based on the density of the fluidized particle, rather than the density of the bed, was plotted against $0.0558 D_p (V - V_{mf}) / V_{mf}$ (see Figure 35). This correlation neglects change in fluidized-bed density with increased gas velocity. The scatter is just slightly more than for the correlation for thermal diffusivity on Figure 34. The correlation in Figure 35 permits an estimation of thermal conductivity from the properties

of the fluidized particles and fluidizing gas and the size of the fixed packing. The correlation on Figure 35 can be expressed as

$$\frac{k_e}{\rho C_p} = \alpha' = (0.0075) 0.0558 D_p \left(\frac{V - V_{mf}}{V_{mf}} \right), \quad (40)$$

where ρ is the density of the fluidized particle. All units are in feet and seconds.

Figure 35

GENERAL CORRELATION FOR THERMAL
DIFFUSIVITIES NEGLECTING DENSITY CHANGES
BECAUSE OF BED EXPANSION

108-7642

The minimum fluidization velocities can be predicted, by Rowe's⁽⁴⁴⁾ method, from the density and viscosity of the gas and the size and density of the fluidized particle.

To summarize, the lateral thermal diffusivities are about one-half the lateral mixing diffusivities in a fluidized-packed bed because the transfer of sensible heat from the fluidized particles is not immediate, resulting in a reduced rate of heat transfer. However, the mechanism of thermal conduction is essentially the same as that of the solids mixing. Therefore, in a manner similar to the correlation for the mixing diffusivities, the thermal diffusivities were correlated as a function of fixed packing diameter and minimum fluidization velocity.

B. Axial Effective Thermal Conductivity

Because the predominant movement of fluidized particles is in the axial direction, the thermal conductivity in the axial direction would be much greater than the thermal conductivity in the lateral direction. It was assumed, in measurement of lateral effective bed thermal conductivities for fluidized-packed beds that the longitudinal temperature gradient was negligible. Lewis *et al.* (28) found that the longitudinal conductivities for the fluidized beds ranged from 100 to 25,000 Btu/(hr)(ft²)(°F/ft). The lateral conductivity for the same fluidized beds never exceeded 2% of the longitudinal conductivity.

Although the longitudinal conductivities are much greater than the lateral conductivities, there is still a finite effective bed thermal conductivity in the longitudinal direction. To have a more accurate estimate of the magnitudes of longitudinal temperature gradients that could be expected in a processing reactor, and the degree of control of the longitudinal temperature gradients, the thermal conductivity of fluidized-packed beds in the axial direction was measured.

1. Apparatus and Procedure

The apparatus for studying longitudinal heat transfer in a fluidized-packed bed is shown in Figure 36. It consisted of a 40-in. column, made from 3.75-in.-ID, stainless-steel tubing. A sintered, stainless-steel disk served as the gas distributor. Air was used as the fluidizing gas and was supplied to the column through the bottom of a hollowed-out disk of 1-in. thick Lucite. This Lucite disk was bolted to a brass flange, which was silver-brazed to the bottom of the column, and the sintered stainless steel disk was held between the Lucite disk and the brass flange. The outer edge of the sintered disk was sealed by high-temperature neoprene gaskets.

The air rate was measured by a rotameter having a range of 0 to 4.3 ft³/min at 70°F and 14.7 psig. This was equivalent to a superficial linear velocity in the column of from 0 to 1.03 ft/sec.

Figure 36
COLUMN FOR MEASURING LONGITUDINAL
THERMAL CONDUCTIVITY IN A
FLUIDIZED-PACKED BED

The heat was introduced and removed internally. A 300 W, 115 V, resistance strip heater, bent to form a hollow cylinder, was placed about 2 in. above the sintered plate. A voltmeter and ammeter indicated the heat input. A 1/4-in. copper coil, through which cooling water flowed, was mounted 14 to 17 in. above the distributor plate. Water flow rates were determined by collecting the water in a one-liter graduated cylinder during a known time interval. Temperature probes were made by drilling holes in 3/8-in.-long by 3/8-in.-diameter brass cylinders, or in 1/2-in.-diameter steel spheres, which were used for the fixed packing, and soft-soldering the end of an iron-constantan thermocouple inside the hole. Ten probes were made for each type of packing material. Eight probes were placed at various longitudinal distances between the heater and the cooling coil. One probe was placed directly on top of the sintered plate, and the other probe was placed at the top of the column to measure the outlet temperature of the air. Thermocouples were also mounted in the inlets and outlets of the water-cooling coils. The entire column was lagged with 2-in. thick asbestos insulation to minimize heat losses to the ambient air.

The fluidized material was No. 070 and No. 120 glass beads and -56 +100 mesh fused alumina grain. Size distribution for these materials as determined by screening are given in Table V. A 3/16-in., stainless-steel tubing, inserted in a hole in the center of the gas distributor, was used to drain the fluidized particles from the column without disturbing the fixed packing. As previously mentioned, 3/8 x 3/8-in. brass cylinders and 1/2-in.-diameter steel spheres were used as the fixed packing.

Table V
FLUIDIZED PARTICLE SIZE DISTRIBUTIONS

Material	Screen Size, Mesh	Weight Percent
No. 070 glass beads	+40	53.8
	+100 -40	45.8
	+120 -100	0.2
	+140 -120	0.1
	-140	0.1
No. 120 glass beads	+40	0.3
	+100 -40	1.4
	+120 -100	94.2
	+140 -120	3.6
	+170 -140	0.4
	-170	0.1
56/100 mesh alumina	+40	0.1
	+100 -40	66.6
	+120 -100	20.3
	+140 -120	12.5
	-140	0.5

2. Discussion of Results

If the thermal conductivity is constant, and if the heat is conducted through a constant cross section, the rate of heat transfer can be expressed as

$$q = k A_s \frac{\Delta T}{\Delta x}, \quad (41)$$

where

q = Heat flux, Btu/hr;

k = Thermal conductivity, Btu/(hr)(ft²)(°F/ft);

A_s = Cross-sectional area, ft²;

T = Temperature, °F;

and

x = Distance, ft.

A typical, longitudinal temperature profile for these runs is shown graphically in Figure 37 as temperatures in °C versus column height in inches. The thermal conductivities for the various combinations of fluidized particles and fixed packing that were studied during this investigation are shown in Figures 38 and 39. The thermal conductivities

Figure 37

LONGITUDINAL TEMPERATURE PROFILE IN A FLUIDIZED-PACKED BED

Figure 38
LONGITUDINAL THERMAL
CONDUCTIVITIES FOR
DIFFERENT FLUIDIZED
MATERIAL

Figure 39
LONGITUDINAL THERMAL
CONDUCTIVITIES FOR
DIFFERENT SHAPES OF
PACKING MATERIAL

shown in these figures are on the basis of the heat transport by the fluidized particles. This was determined by the heat pickup of the cooling water. The contribution to the total heat flux by the air stream was neglected, except as described below, and the radial temperatures were assumed uniform.

Tables VI through IX list the contributions to the total heat flux by the air stream and the fluidized particles. The amounts of heat transported

Table VI

EXPERIMENTAL DATA FOR 2000 g OF NO. 070 GLASS BEADS
IN A 19-in.-DEEP BED OF 3/8 x 3/8-in. BRASS CYLINDERS

Run No.	G, cfm (70°F, 14.7 psia)	V ₀ , Superficial Air Velocity, ft/sec	$\frac{\Delta T}{\Delta x}$, °F/ft	Q (air), Btu/hr	Q (cooling), Btu/hr	k, Btu/ (hr)(ft²) (°F/ft)
52	0	0	76.8	0	17	2.9
51	0.4	0.096	38.0	7	131	44.4
50	0.8	0.191	37.0	11	166	58.0
49	1.3	0.311	26.9	22	182	87.2
41	1.3	0.311	33.3	39	186	72.0
42	1.7	0.406	36.0	36	186	66.4
43	2.1	0.502	28.5	45	201	91.2
40	2.6	0.621	21.2	81	233	142.0
44	2.6	0.621	22.0	61	210	123.2
45	3.0	0.717	21.6	74	219	130.8
46	3.4	0.812	23.8	87	240	130.4
47	3.9	0.931	24.4	152	252	133.2
48	4.3	1.027	28.2	166	268	122.8

Table VII

EXPERIMENTAL DATA FOR 2000 g OF NO. 120 GLASS BEADS
IN A 19-in.-DEEP BED OF 3/8 x 3/8-in. BRASS CYLINDERS

Run No.	G, cfm (70°F, 14.7 psia)	V ₀ , Superficial Air Velocity, ft/sec	$\frac{\Delta T}{\Delta x}$ °F 'ft	Q (air), Btu/hr	Q (cooling), Btu/hr	k, Btu/ (hr)(ft ²) (°F/ft)
61	0	0	75.5	0	8	1.4
57	0.4	0.096	18.1	3	280	202
56	0.8	0.191	14.4	8	307	278
55	1.3	0.311	11.0	16	321	380
54	1.7	0.406	10.2	22	325	416
60	1.9	0.454	14.8	29	350	308
53	2.1	0.502	16.0	33	369	300
58	2.1	0.502	16.1	39	346	280
59	2.6	0.621	17.2	42	372	282

Table VIII

EXPERIMENTAL DATA FOR 2000 g of 56/100 MESH ALUMINA
IN A 19-in.-DEEP BED OF 3/8 x 3/8-in. BRASS CYLINDERS

Run No.	G, cfm (70°F, 14.7 psia)	V ₀ , Superficial Air Velocity, ft/sec	$\frac{\Delta T}{\Delta x}$ °F 'ft	Q (air), Btu/hr	Q (cooling), Btu/hr	k, Btu/ (hr)(ft ²) (°F/ft)
74	0	0	69.0	0	19.4	3.7
67	0.4	0.096	34.2	8	182	69
66	0.8	0.191	33.3	16	204	80
67	1.3	0.311	29.2	28	227	101
64	1.3	0.311	32.4	36	209	84
63	1.7	0.406	29.4	52	233	103
62	2.1	0.502	29.5	58	246	109
73	2.3	0.550	27.4	65	274	130
68	2.6	0.621	24.5	74	279	148
69	3.0	0.717	25.3	86	285	147
70	3.4	0.812	29.5	100	317	140
71	3.9	0.931	31.8	123	321	132
72	4.3	1.027	32.5	142	327	132

Table IX

EXPERIMENTAL DATA FOR 2000 g OF 56/100 MESH ALUMINA
GRAIN IN A 17-in.-DEEP BED OF 1/2-in. -diam STEEL SPHERES

Run No.	G, cfm (70°F, 14.7 psia)	V ₀ , Superficial Air Velocity, ft/sec	$\frac{\Delta T}{\Delta x}$ °F 'ft	Q (air), Btu/hr	Q (cooling), Btu/hr	k, Btu/ (hr)(ft ²) (°F/ft)
92	0	0	220	0	15.2	0.90
83	0.4	0.096	20.7	15	309	194
82	0.8	0.191	15.6	29	329	274
81	1.3	0.311	12.4	47	324	340
80	1.7	0.406	8.8	61	350	519
91	1.9	0.454	7.5	70	328	569
90	2.1	0.502	7.5	78	328	574
84	2.1	0.502	6.8	81	328	628
85	2.6	0.621	7.7	98	338	571
86	3.0	0.717	8.8	111	338	500
84	3.4	0.812	9.0	120	326	472

to the cooling water by the fluidized particles and by the air were determined from the fluid flow rates, their heat capacities, and their change in temperature. These quantities appear as Q (air) and Q (cooling). The contribution of heat transfer by the air stream became significant at higher velocities. At about 1 ft/sec the air carried away about 40% as much heat as the water. Figure 40 is a plot of thermal conductivity based on Q (cooling) plus Q (air) versus gas velocity. The thermal conductivities based on the total heat flux (heat transport by particles and air) is greater than the heat flux just based on the transport by the fluidized particles.

Figure 40
LONGITUDINAL THERMAL CONDUCTIVITIES FOR
FLUIDIZED-PACKED BEDS, BASED ON
TOTAL HEAT FLUX

108-7340

The curves in Figures 38 and 39, which were calculated on the basis of Q (cooling) alone, have a maximum between 0.4 and 0.7 ft/sec because of the interaction of two effects. The thermal conductivity of a fluidized-packed bed depends both on the movement of the fluidized particles and on their density. As the gas velocity increases, the particle motion increases. But at the same time, increasing the velocity expands the bed of particles and decreases the particle density. This decrease in density compensates for the effect of the increased particle motion and the conductivity attains a maximum, and then the particle density effect

predominates with further increase in gas velocity causing a decrease in the effective bed thermal conductivity. Also the particle motion may attain a maximum with increased gas velocity as shown in Sect. III-B on axial solids mixing.

Figure 38 shows the strong dependence of conductivity on packing shape. Spheres pack more uniformly and have a greater void space than do cylinders. This increases the freedom of motion for the fluidized particles, which results in a higher conductivity for a bed of spheres. The difference in conductivities is about a factor of four between the 1.2-in. steel spheres and the $3/8 \times 3/8$ -in. brass cylinders.

3. Conclusions

In the present investigation, values of up to 600 Btu/(hr)(ft²)(°F/ft) were obtained for the longitudinal thermal conductivities in fluidized-packed beds, and these values appear to be a factor of more than 20 higher than lateral thermal conductivities obtained in similar systems in previous investigations. However, because of the limited number of variables studied, it is not possible to describe a general correlation for longitudinal thermal conductivities, as was done for lateral thermal conductivities.

C. Heat-transfer Coefficients

1. Heat-transfer Coefficients at the Walls of a Fluidized-packed Bed

Wall heat-transfer coefficients were measured with a 2.88-in.-ID and a 3.88-in.-ID column. Heat was supplied by $3/8$ -in.-diameter heaters, axially mounted in the columns, and heat was removed from the column walls by water cooling. Porous metal was used as the gas distributor.

a. 2.88-in.-ID Column

For the runs with the 2.88-in. column, glass beads (average diameter 0.0045 in.) were fluidized with air in the voids of a 28-in. deep bed of $3/8 \times 3/8$ -in. brass cylinders. The active length of the axially mounted heater was 27 in.

Figure 41 describes the variation of heat-transfer coefficients at the heater and at the cooling wall for this system with increasing gas velocity. The average gas temperature in the column was about 212°F. The data for the fluidized-packed bed were compared with predicted heat-transfer coefficients for a fluidized bed of 0.0045-in. glass beads. Comparisons were made with the correlation by Wen and Leva⁽⁵⁴⁾ for heat-transfer coefficients at the cooling wall, and by Vreendenberg⁽⁵²⁾ for heat-transfer coefficients at the internal heater. The heat-transfer coefficients for the fluidized-packed bed are somewhat lower than the

heat-transfer coefficients obtained from the generalized correlation for a fluidized bed. This would be expected because of the baffled solids flow in the fluidized-packed bed.

Figure 41
FLUIDIZED-PACKED-BED HEATER
AND WALL FILM COEFFICIENT,
 h , AS A FUNCTION OF GAS
VELOCITY

The heat-transfer coefficients at the heater wall are greater than at the cooling wall. This would be expected from an examination of the equation developed by Ziegler *et al.*⁽⁶¹⁾ for relating the wall heat-transfer coefficient to the particle residence time at the wall surface:

$$\frac{hd}{k_g} = \frac{4\pi/3}{\left[1 + \frac{3k_g \bar{\theta}}{\rho_s C_s R_p^2 (\alpha_s + 1)} \right]^{\alpha+1}}, \quad (42)$$

where

- h = Wall heat-transfer coefficient, Btu/(hr)(ft²)(°F);
- d = Fluidized particle diameter, ft;
- k_g = Thermal conductivity of gas, Btu/(hr)(ft²)(°F/ft);
- $\bar{\theta}$ = Mean particle residence time, hr;
- ρ_s = Particle density, lb/ft³;
- C_s = Particle heat capacity, Btu/(lb)(°F);
- R_p = Particle radius, ft;

and

α_s = Shape factor for gamma distribution.

Particles moving near the small internal heater have more directions of escape than the particles near the column wall, which are bounded in one direction by the wall. Therefore, the residence time of the particles near the internal heater would be less than for the particles at the column wall. From Equation 42, it would be anticipated that the heater heat-transfer coefficients would be greater than the wall heat-transfer coefficients since the heat-transfer coefficient, h , is an inverse function of the mean particle residence time, $\bar{\theta}$. Also, the bubble frequency and hence, the rate of particle movement, may be less at the column wall than at the center.

b. 3.88-in.-ID Column

Nitrogen was the fluidizing gas, and 3/8-in.-diameter steel spheres were the packing for all runs in the 3.88-in.-ID column. Three sizes of copper-nickel shot were fluidized: -40 +50, -100 +120, and -140 +170 mesh. The length of the axially located 3/8-in.-diameter heater was 16 in. The bed depth for each run was also 16 in. The quantity of fluidized particles was adjusted so that the expanded fluidized-bed portion was always at the same height as the fixed packing.

Figure 42 shows the heat-transfer coefficients at the wall, and at the heater, for the various fluidized particle sizes. For the runs with the -100 +120 and -140 +170 mesh particles, the column walls were cooled with water. The column was air-cooled for the run with the

Figure 42
HEATER AND WALL HEAT TRANSFER
COEFFICIENTS FOR DIFFERENT SIZES OF
COPPER-NICKEL SHOT FLUIDIZING IN THE
VOIDS OF 3/8-IN. SPHERICAL PACKING

-40 +50 mesh copper-nickel particles resulting in a low heat flux. Because of a low temperature difference between the wall and the bed interior for the case of the -40 +50 mesh copper-nickel particles, the calculated wall heat-transfer coefficients were very erratic and are, therefore, omitted from the figure.

As for the 2.88-in. column with glass beads and $3/8 \times 3/8$ -in. cylindrical packing, the wall heat-transfer coefficients are lower than the heat-transfer coefficients at the heater. A comparison of the heater heat-transfer coefficients for the different particle sizes shows an increase in heat-transfer coefficient with decreasing particle size.

In summary, the heat-transfer coefficients for a fluidized-packed bed are up to a factor of two less than for the same fluidized particles in an open tube. The heat-transfer coefficients are lower at the column wall than at the internal heater wall.

2. Heat-transfer Coefficients from the Surface of Fixed Packing in a Fluidized-packed Bed

The heat-transfer coefficients from the surface of the fixed packing were measured for a fluidized-packed bed consisting of brass cylinders, $1/2$ in. in diameter by $3/4$ in. long, and of -140 +200 mesh copper shot. Nine electrical resistance heaters, each of the same dimensions as the brass cylindrical packing, were dispersed in the fixed packing over a 2.5-in. height. The orientation of the heaters and packing was random.

The pellet bed height was held constant at 10.7 in. in these tests, and the static height of the fluidized medium was varied from 5.8 to 12.7 in. The heated portion of the bed was always immersed. Measurements were made in a 2.88-in.-ID column with a porous-metal gas distributor.

Iron-constantan thermocouples were welded to the surface of three heaters and six brass cylinders. The heat-transfer coefficients shown in Figure 43 were calculated as a single value based on the difference of the average temperature of the heaters and the average temperature of the brass cylinders. The heaters were in series, and therefore the current to each heater was the same. The heat output for each cylindrical heater was varied from 85 to 102 Btu/hr. This heating rate is of the same order as may be encountered in the fluorination of uranium dioxide pellets. For comparison, heat-transfer coefficients for the packed bed without the fluidizing particles are also shown in Figure 43.

Figure 43
FIXED PACKING SURFACE HEAT
TRANSFER COEFFICIENTS IN A
FLUIDIZED-PACKED BED

108-4539

The above data for average heat-transfer coefficients did not indicate any dependence on the height of the fluidized particles in the range studied.

To indicate the variation of heat-transfer coefficients for individual pellet heaters, heat-transfer coefficients were calculated on the basis of the difference between the temperature of a particular pellet heater and the average temperature of three brass pellets nearest the pellet heater. The variation in the individual heater heat-transfer coefficients were quite large, depending on differences of turbulence and circulation of the fluidized particles about the particular orientation of the fixed pellets. As can be seen from Figure 44, these variations are smaller when the gas velocity is several times the minimum fluidization velocity.

There is also the possibility of high, localized, heat transfer by conduction with adjoining brass cylinders. If the flat surfaces of a cylindrical pellet heater and an adjacent brass pellet were in contact, heat transfer by conduction between the pellets would be high in the area of contact. However, in the correlation by Yagi and Kunii⁽⁵⁷⁾ for packed-bed thermal conductivities, no distinction is made between cylindrical and spherical packing, and as pointed out in Sect. IV-A on lateral thermal conductivity, the contribution by packing conductivity is negligible, and heat transport is essentially by the fluidized particles.

These tests have indicated that the heat-transfer coefficient at the pellet surface in a packed bed is increased by a factor of about eight with the addition of fluidized particles in the voids of the fixed packing.

Figure 44

HEAT-TRANSFER COEFFICIENTS FOR
THREE INDIVIDUAL PELLET HEATERS
IN A FLUIDIZED-PACKED BED

108-4543

V. GAS MIXING

Process control is dependent on reactant gas distribution as well as heat removal rates. To predict mass gradients in the reactor, gas diffusivities must be determined. Knowledge of the rates of gas diffusion is necessary in the design of gas injectors.

Gas diffusivities have been reported for the separate systems of fluidized beds and packed beds, but the gas-mixing behavior in the combined fluidized-packed bed has remained unknown.

A. Fluidized Beds

The main emphasis of previous work on fluidized beds has been concerned with gas back-mixing.^(1,3,19,23,36,42) Back-mixing of gas was reduced by using baffles and by selecting fluidized materials that have a low gas-absorption capacity. Baerns *et al.*⁽²⁾ measured radial as well as longitudinal gas mixing in fluidized beds. Using brown nitrogen dioxide as a tracer gas in a transparent column, Wace and Burnett⁽⁵³⁾ found that, for gas flows slightly above the minimum fluidization velocity, the gas flow appeared streamlined in spite of violent solids movement caused by the passing bubbles. Although distorted and displaced by the pressure gradients associated with the bubbles, the tracer gas streams persisted as coherent threads. In an analysis of gas back-mixing in fluidized beds, Rowe⁽⁴³⁾ attributed the spreading of tracer gas concentrations to variations of residence times in the bed for the gas flow elements. These variations are caused by bubble disturbances of the gas flow.

B. Beds of Fixed Packing

Although little work has been reported in the literature on radial mass transfer in fluidized beds, a considerable amount has been reported on beds with fixed packing. Bernard and Wilhelm⁽⁷⁾ presented the first paper on mass transfer within the fluid interstices of the packing. They used both gas and liquid systems. Singer and Wilhelm⁽⁴⁷⁾ then compared convective transport of heat in a packed bed with the eddy diffusivities obtained from the mass-transfer measurements and obtained reasonable agreement. Plautz and Johnstone⁽³⁷⁾ did further work using sulfur dioxide as a tracer gas. Fahien and Smith⁽¹⁵⁾ obtained gas diffusivities for various radial positions. Dorweiler and Fahien⁽¹⁴⁾ later extended this work to regions of low gas flow.

For packed beds, gas eddy diffusivity, E , is determined by the packing diameter, D_p , and the gas velocity, V . For correlations, dimensionless groups are used, namely the Peclet number, $D_p V/E$, which contains the diffusivity terms, and the Reynolds number, $D_p V_0 \rho/\mu$,

which contains the gas properties (density, ρ , and viscosity, μ). Considering the packed particles as points of deflection, Baron⁽⁴⁾ determined the mean free path of elements of the gas stream and thence the turbulent diffusion coefficient by means of a random-walk calculation. He predicted the Peclet number should be between 5 and 13 for packed beds. Latinen,⁽²⁵⁾ using a similar approach for a body-centered, cubic, packing arrangement, predicted a Peclet number of 11.3. Ranz⁽³⁸⁾ gave a Peclet number of 11.2 for a system of packing with the centers at the corner of tetrahedrons. These methods assumed spherical particles and a uniform void space, and agree with experimental results for a low particle-to-tube diameter ratio.

This paper reports radial gas mixing for various fluidizing and packing materials measured by using carbon dioxide as a tracer gas in a stream of nitrogen. Using the mathematical method developed by Hiby and Schümmer,⁽²⁴⁾ eddy diffusivities were measured. The radial mixing was essentially determined by the size of the fixed packing in the fluidized-packed bed.

C. Theory

Eddy diffusivity, E , can be defined by the following equation:

$$N = -(D_v + E) \frac{dC}{dy}, \quad (43)$$

where N is the rate of mass transfer, D_v is the molecular diffusivity, and C is the gas concentration.

At steady state, the partial differential equation in cylindrical coordinates for diffusion-convection is

$$V \frac{\partial C}{\partial z} = E_t \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} + \frac{\partial^2 C}{\partial z^2} \right). \quad (44)$$

Equation 44 assumes that the total diffusivity, E_t , in the radial direction equals E_t in the longitudinal direction, and that E_t and V , the point velocity of the gas, do not vary with positions in the bed. V can then be equated to the superficial gas velocity divided by the void fraction. Wilson⁽⁵⁶⁾ solved this equation for the case of no column wall boundary (infinite diameter) and bed-height-to-radius ratio, z/R , greater than 5. Written for a mass-transfer situation in which a tracer material is released at a point, the equation is

$$\frac{C}{C_a} = \frac{V_0 R^2}{4 E_t z} \exp \left(\frac{V_0 r^2}{\epsilon E_t 4 z} \right). \quad (45)$$

A plot of $\ln(C/C_a)$ versus r^2 yields a linear plot of slope $(-V_0/\epsilon E_t 4 z)$. By this means, the total diffusivity can be obtained from an

experimentally determined, radial-concentration profile. This method was used by Bernard and Wilhelm⁽⁷⁾ and by Plautz and Johnstone.⁽³⁷⁾ However, it requires obtaining concentration samples at several points along the column diameter, and is time-consuming. Another method was developed by Hiby and Schümmer⁽²⁴⁾ as a solution of Equation 44 for a tracer gas injector tube of same radius as the gas sample tube. Both tubes are of finite radius concentric in a column of a finite radius. Only one sample has to be taken to determine the total diffusivity, E_t . Hiby and Schümmer's solution is

$$\frac{\bar{C}}{C_0} = 4 \sum_{n=0}^{\infty} \frac{J_1^2 \lambda_n A_r / R}{\lambda_n^2 J_0^2(\lambda_n)} \exp \left(-\frac{z}{D_p} \frac{\lambda_n^2}{K^2} \frac{E_t}{D_p V} \right) \quad (46)$$

where $\bar{C}$ = average gas concentration in sample tube. If the ratio of injector-tube radius to column radius, A_r/R , approaches 0, then Equation 4 simplifies to

$$\frac{\bar{C}}{C_0} = \frac{1}{4} \sum_{n=0}^{\infty} (-1)^n \frac{(2n+2)!}{2^{2n}(n+2)(n+1)!^3} \left(\frac{A_r^2 D_p V}{D_p z E_t} \right)^{n+1}. \quad (47)$$

Equations 46 and 47 are described graphically on Figure 45.

Experimental comparison of the two methods gave results that were in reasonable agreement. For the work reported in this paper, the method developed by Hiby and Schümmer was used for determining the diffusivities.

Peclet numbers, $D_p V / E_t$, were initially determined experimentally in terms of total diffusivity, and Peclet numbers for eddy diffusivity were then calculated. The eddy diffusivity, E , was found by subtracting the molecular diffusivity, D_v , from the total diffusivity. The molecular diffusion coefficient for carbon dioxide in nitrogen is 0.612 ft²/hr at 20°C.⁽¹⁸⁾ The mean point velocity, V , was calculated by assuming a void fraction, ϵ , of 0.4 for the fixed packing, and dividing the void fraction into the superficial gas velocity, V_0 .

For determining the effect of fluidized material in the voids of the fixed packing on the gas mixing, Peclet numbers were based on the diameter of the fixed packing, and Reynolds numbers were based on the properties of the gas (density and viscosity) as well as the fixed-packing diameter. This allows comparison with eddy diffusivity data reported in the literature for packed beds.

D. Equipment and Experimental Procedure

A schematic diagram of the column used for eddy diffusion measurements is shown in Figure 46. The column was made from a 4-in. copper tube (3.875-in. ID), 34 in. long. The bed was supported on a sintered metal

Figure 46
SCHEMATIC DIAGRAM OF
COLUMN USED FOR EDDY
DIFFUSION MEASUREMENTS

Figure 45
HIBY AND SCHÜMMER RELATION-
SHIP FOR SAMPLE CONCENTRA-
TION AND DIFFUSIVITY

plate. Beneath the support plate was a 6-in. calming section, consisting of 1/4-in. glass beads. Nitrogen was introduced at the base of the calming section, and tracer gas (carbon dioxide) was introduced through a 1/4-in. nickel tube (0.19-in. ID) which extended 2 in. above the sintered metal support plate. The tracer gas injector tube extended above the base of bed to reduce end effects. A screen was placed over the tip of the injector tube to prevent packing from blocking the gas flow. A sample probe was also fabricated of 1/4-in. nickel tubing and was movable to permit gas sampling at various radial positions. The probe was mounted so that the sample point was 12 in. above the point of tracer gas input. The sampler tube opening was covered with a piece of cotton cloth to prevent the entrance of fluidized particles.

Various sizes and shapes of fixed packing and fluidized particles were used. The fixed packing consisted of 1/8-, 1/4-, and 1/2-in. steel spheres and 3/8 x 3/8-in. brass cylinders. Glass beads with average diameters of 0.0110 and 0.0080 in., -100 +140 mesh copper shot, and -54 +100 mesh Alundum were used for fluidized materials.

The gas flow rates were monitored with rotameters and regulated so that the velocities in the sample and injector tubes were the same as the velocity of the main gas stream. Tracer gas concentrations were determined by gas thermal conductivity measurements. A mixture of 2.00% carbon dioxide in nitrogen prepared by the Matheson Chemical Company was used as the standard for calibrating the conductivity cell.

The method described by Hiby and Schümmer was used for determining the diffusivities rather than the one used by Bernard and Wilhelm, because just one gas sample was necessary. A comparison was made using the two methods for a run with 1/2-in. steel balls, no fluidized bed, and a superficial gas velocity of 1.0 ft/sec.

Figure 47 is the experimental radial concentration profile for the above conditions. From a plot of $\ln(C/C_a)$ versus r^2 (see Figure 48) the Peclet number can be obtained in the following manner:

$$\text{Slope of } \ln(C/C_a) \text{ versus } r^2 = -0.5 \text{ in.}^{-2},$$

or

$$-\frac{V_0}{\epsilon E_t 4z} = -0.5 \text{ in.}^{-2},$$

$$\frac{D_p V}{E_t} = (0.5)(144)(1/24)(4)(1) = 12.0.$$

Figure 47
 CONCENTRATION PROFILE OF
 TRACER GAS IN A PACKED BED

Figure 48
 SEMILOG PLOT OF CONCENTRATION
 AS A FUNCTION OF r^2

The Peclet number can be obtained from just the concentration at $r = 0$ by using the method of Hiby and Schümmer:

$$\text{For } \frac{\bar{C}}{C_0} = 0.0046, \frac{A_r^2 D_p V}{D_p z E_t} = 0.0185 \text{ (see Figure 45)}$$

$$\frac{D_p V}{E_t} = (0.0185)(144)(1/2 \times 1/12 \times 1)/(0.19/2)^2 = 12.2.$$

Both methods are in reasonable agreement. Because the Hiby and Schümmer method requires less samples, it was used in all of the runs.

E. Experimental Results

1. Packed Bed

Figure 49 compares the results obtained for a packed bed of 1/2-in. spheres and some literature values. There is a fair agreement, indicating that the experimental technique was satisfactory.

Figure 49

PECLET NUMBER AS A FUNCTION OF GAS VELOCITY FOR PACKED BEDS

The data for all the runs are tabulated in Tables X-XV.

Table X
 PECLET NUMBERS FOR 1/2-in. SPHERICAL PACKING

Run	Fluidizing Material	V_0 , ft/sec	$D_p V_0 \rho / \mu$	Concentration % CO ₂	$\frac{A_p^2 D_p V}{D_p z E_t}$	$D_p V / E_t$	E_t , ft ² /hr	$E_t - 0.61$	$D_p V / E$
ED-4	None	0.3	80.1	0.56	0.0225	14.9	7.57	6.96	16.2
		0.5	133.8	0.56	0.0225	14.9	12.6	12.0	15.6
		0.7	187.2	0.51	0.0205	13.6	20.1	19.5	14.0
		1.0	268	0.46	0.0185	12.2	30.9	30.3	12.4
		1.4	375	0.41	0.0165	10.9	48.3	47.7	11.0
ED-5	Glass beads, average diam 0.0110 in.	0.3	80.1	0.52	0.2100	13.9	8.12	7.51	15.0
		0.5	133.8	0.53	0.0213	14.1	13.3	12.7	14.8
		0.7	187.2	0.55	0.0220	14.6	18.7	18.1	15.1
		1.0	268	0.50	0.0200	13.2	28.6	28.0	13.5
		1.4	375	0.51	0.0250	13.6	38.7	38.1	13.8
ED-19	Glass beads, average diam 0.0110 in.	0.3	80.1	0.42	0.0168	11.1	10.2	9.6	11.8
		0.5	133.8	0.45	0.0180	11.9	15.8	15.2	12.4
		0.7	187.2	0.52	0.0210	13.9	19.6	19.0	14.4
		1.0	268	0.45	0.0180	11.9	31.7	31.1	12.1
ED-11	Glass beads, average diam 0.0080 in.	0.3	80.1	0.32	0.0120	8.0	14.1	13.5	8.4
		0.5	133.8	0.50	0.0200	13.2	14.2	13.6	13.8
		0.7	187.2	0.71	0.0287	19.0	14.4	13.8	19.8
		1.0	268	0.61	0.0245	16.2	23.3	22.7	16.6
		1.4	375	0.58	0.0233	15.4	34.2	33.6	15.7
ED-15	+54 -100 mesh Alundum	0.3	80.1	0.81	0.0327	21.6	5.22	4.61	24.5
		0.5	133.8	0.56	0.0225	14.9	12.6	12.0	15.7
		0.7	187.2	0.60	0.0241	16.0	17.0	16.4	16.6
		1.0	268	0.48	0.0193	12.8	29.4	28.8	13.1
		1.4	375	0.38	0.0152	10.1	52.1	51.5	10.2
ED-18	None	0.3	80.1	0.61	0.0245	16.2	6.96	6.35	17.8
		0.5	133.8	0.62	0.0250	16.6	11.3	10.7	17.5
		0.7	187.2	0.59	0.0220	14.6	18.7	18.1	15.1
		1.0	268	0.49	0.0196	13.0	29.0	28.4	13.3
		1.4	375	0.47	0.0188	12.5	42.1	41.5	12.7

Table XI
 PECLET NUMBERS FOR 1/4-in. SPHERICAL PACKING

Run	Fluidizing Material	V_0 , ft/sec	$D_p V_0 \rho / \mu$	Concentration % CO ₂	$\frac{A_p^2 D_p V}{D_p z E_t}$	$D_p V / E_t$	E_t , ft ² /hr	$E_t - 0.61$	$D_p V / E$
ED-6	None	0.3	40.1	0.90	0.0365	12.1	4.65	4.04	13.9
		0.5	66.8	0.95	0.0383	12.7	7.38	6.77	14.0
		0.7	93.7	0.96	0.0387	12.9	10.2	9.6	13.7
		1.0	134	0.87	0.0350	11.6	16.2	15.6	12.0
		1.4	187.5	0.81	0.0327	10.9	24.1	23.5	11.2
ED-7	Glass beads, average diam 0.0110 in.	0.3	40.1	0.9	0.0363	12.1	4.65	4.04	13.9
		0.5	66.8	0.85	0.0343	11.4	8.23	7.62	12.3
		0.7	93.7	1.12	0.0453	15.1	8.70	8.09	16.2
		1.0	134	0.83	0.0334	11.1	16.9	16.3	11.5
		1.4	187.5	0.60	0.0240	7.97	32.9	32.3	8.1
ED-8	Glass beads, average diam 0.0080 in.	0.3	40.1	0.34	0.0131	4.35	12.9	12.3	4.6
		0.5	66.8	0.46	0.0185	6.15	15.3	14.7	6.4
		0.7	93.7	0.47	0.0188	6.25	21.0	20.4	6.4
		1.0	134	0.38	0.0152	5.05	37.1	36.5	5.2
		1.0	134	0.65	0.0261	8.67	21.6	21.0	8.9
		1.4	187.5	0.63	0.0253	8.40	31.2	30.6	8.6

Table XII
 PECLET NUMBERS FOR 1/8-in. SPHERICAL PACKING

Run	Fluidizing Material	V_0 , ft/sec	$D_p V_0 \rho / \mu$	Concentration % CO ₂	$\frac{A^2 D_p V}{D_p z E_t}$	$D_p V / E_t$	E_t , ft ² /hr	$E_t - 0.61$	$D_p V / E$
ED-13	None	0.3	20.0	1.58	0.0641	10.7	2.64	2.03	13.9
		0.5	33.4	1.63	0.0662	11.0	4.26	3.65	12.8
		0.7	46.9	1.75	0.0712	11.8	5.56	4.95	13.3
		1.0	67.0	1.45	0.0590	9.80	9.56	8.95	10.5
		1.4	93.8	1.31	0.0532	8.84	14.85	14.24	9.2
ED-14	Glass beads, average diam 0.0080 in.	0.3	20.0	0.55	0.0220	3.65	7.73	7.12	4.0
		0.5	33.4	0.78	0.0315	5.23	8.97	8.66	5.4
		0.5	33.4	1.35	0.0548	9.10	5.15	4.54	10.3
		0.7	46.9	0.70	0.0281	4.66	14.08	13.47	4.9
		1.0	67.0	1.26	0.0510	8.45	11.10	10.49	8.9
ED-16	Glass beads, average diam 0.0110 in.	1.4	93.8	1.00	0.0404	6.70	19.59	18.98	6.9
		0.3	20.0	1.11	0.0450	7.46	3.78	3.17	8.9
		0.5	33.4	1.07	0.0432	7.16	6.55	5.94	7.9
		0.7	46.9	1.21	0.0490	8.15	8.05	7.44	8.8
		1.0	67.0	1.13	0.0458	7.61	12.31	11.70	8.0
ED-17	Glass beads, average diam 0.0110 in.	1.4	93.8	1.14	0.0461	7.65	17.25	16.64	7.9
		0.3	20.0	0.67	0.0270	4.48	6.30	5.69	5.0
		0.5	33.4	0.79	0.0319	5.30	8.85	8.24	5.7
		0.7	46.9	0.95	0.0382	6.35	10.33	9.72	6.8
		1.0	67.0	0.97	0.0390	6.48	14.45	13.84	6.8

Table XIII
 PECLET NUMBERS FOR 3/8-in. CYLINDRICAL PACKING

Run	Fluidizing Material	V_0 , ft/sec	$D_p V_0 \rho / \mu$	Concentration % CO ₂	$\frac{A^2 D_p V}{D_p z E_t}$	$D_p V / E_t$	E_t , ft ² /hr	$E_t - 0.61$	$D_p V / E$
ED-2	None	0.3	60.1	0.37	0.0148	7.36	11.8	11.2	7.8
		0.4	80.3	0.37	0.0148	7.36	15.8	15.2	7.6
		0.5	100.2	0.36	0.0142	7.07	20.5	19.9	7.3
		0.6	120.5	0.37	0.0148	7.36	23.6	23.0	7.6
		1.0	201	0.34	0.0130	6.48	44.7	44.1	6.6
		1.4	281	0.32	0.0120	5.98	67.9	67.3	6.0
ED-3	Glass beads, average diam 0.0110 in.	0.3	60.1	0.37	0.0148	7.37	11.8	11.2	7.8
		0.5	100.2	0.35	0.0136	6.78	21.4	20.8	7.0
		1.0	201	0.31	0.0116	5.78	50.2	48.6	6.0
ED-19	Glass beads, average diam 0.0110 in.	0.3	60.1	0.60	0.0241	12.0	7.25	6.64	13.1
		0.5	100.2	0.46	0.0185	9.21	15.7	15.1	9.6
		1.0	201	0.36	0.0142	7.07	41.0	40.4	7.2
		1.4	281	0.26	0.0082	4.08	99.5	98.9	4.1
ED-1	Glass beads, average diam 0.0080 in.	0.3	60.1	0.43	0.0172	8.56	10.2	9.6	9.1
		0.4	80.3	0.47	0.0188	9.35	12.4	11.8	9.8
		0.5	100.2	0.51	0.0210	10.45	13.9	13.3	10.9
		0.6	120.5	0.51	0.0210	10.45	16.7	16.1	10.8
		1.0	201	0.40	0.0160	7.96	36.4	35.8	8.1
		1.4	281	0.40	0.0160	7.96	51.0	50.4	8.1
ED-12	+100 -140 mesh copper shot	0.3	60.1	0.44	0.0187	9.30	9.35	8.74	10.0
		0.5	100.2	0.52	0.0210	10.45	13.9	13.3	10.9
		0.7	140.2	0.53	0.0213	10.6	19.2	18.6	10.9
		1.0	201	0.41	0.0164	8.16	35.5	34.9	8.3
		1.4	281	0.51	0.0205	10.2	39.8	39.2	10.4

Table XIV
PECLET NUMBERS FOR OPEN TUBE

Run	Fluidizing Material	V_0 , ft/sec	$D_p V_0 \rho / \mu$	Concentration % CO ₂	$\frac{A^2 D_t V}{D_t^2 E_t}$	$D_t V / E_t$	E_t , ft ² /hr	$E_t - 0.61$	$D_t V_0 / E$
ED-8A	None	0.3	621	0.42	0.0168	86.5	4.03	3.42	102
		0.5	1038	0.47	0.0188	97.0	6.01	5.40	108
		0.7	1451	0.48	0.0192	99.0	8.24	7.63	107
		1.0	2080	0.47	0.0188	97.0	12.0	11.4	102
		1.4	2910	0.45	0.0180	92.8	17.6	17.0	96
ED-9	Glass beads, average diam 0.0080 in.	0.3	621	0.36	0.0142	73.1	4.77	4.16	84
		0.4	831	0.27	0.0090	46.4	10.0	9.4	50
		0.5	1038	0.25	0.0070	36.1	16.2	15.6	37
		0.7	1451	0.22	0	0	-	-	0
ED-10	Glass beads, average diam 0.0110 in.	0.3*	621	5.83	0.250	1290	0.68**	0.07	12,450
		0.4	831	0.56	0.0225	116	4.02	3.41	137
		0.5	1038	0.37	0.0147	75.8	7.70	7.09	82
		0.7	1451	0.36	0.0142	73.1	11.2	10.6	77
		1.0	2080	0.32	0.0121	62.4	18.7	18.1	64
		1.4	2910	0.25	0.0070	36.1	45.2	44.6	37

*Below minimum fluidization velocity.

**V, point velocity, based on voidage in particles not on open tube.

Table XV
PECLET NUMBERS FOR VARIOUS SYSTEMS
BASED ON COLUMN DIAMETER

Packing	Fluidizing Material	V_0	$D_t V_0 \rho / \mu$	$D_p V / E$	$D_t V / E$
1/2-in. spheres	None	0.3	621	16.2	126
		0.3	621	17.8	138
		0.5	1038	15.6	121
		0.5	1038	17.5	136
		0.7	1451	14.0	108
		0.7	1451	15.1	117
		1.0	2080	12.4	96
		1.0	2080	13.3	103
		1.4	2910	11.0	85
		1.4	2910	12.7	98
1/4-in. spheres	None	0.3	621	13.9	216
		0.5	1038	14.0	217
		0.7	1451	13.7	212
		1.0	2080	12.0	186
		1.4	2910	11.2	174
1/8-in. spheres	None	0.3	621	13.9	430
		0.5	1038	12.8	396
		0.7	1451	13.3	412
		1.0	2080	10.5	325
		1.4	2910	9.2	285
3/8 x 3/8-in. cylinders	None	0.3	621	7.8	81
		0.4	829	7.6	79
		0.5	1038	7.3	75
		0.6	1242	7.6	79
		1.0	2080	6.6	68
		1.4	2910	6.0	62
1/4-in. spheres	0.0110-in. glass beads	0.3	621	13.9	216
		0.5	1038	12.3	191
		0.7	1451	16.2	251
		1.0	2080	11.5	178
		1.4	2910	8.1	126

2. Fluidized-packed Bed

In the runs in which there was fixed packing but no fluidizing particles, the point concentrations of the carbon dioxide tracer gas showed a radial concentration profile that was regular with the peak concentration near the center of the column. These runs without fluidizing material resulted in a plot of Peclet number versus Reynolds number, which is shown by continuous curves in Figures 50, 51, 52 and 53. Other points in these figures are from runs with fluidized material.

Figure 50

PECLET NUMBERS FOR
FLUIDIZED-PACKED BED PACKED
WITH 1/2-in. SPHERES

108-5868 Rev.

Figure 51

PECLET NUMBERS FOR
FLUIDIZED-PACKED BED PACKED
WITH 1/4-in. SPHERES

108-5863 Rev.

Figure 52

PECLET NUMBERS FOR
FLUIDIZED-PACKED BED PACKED
WITH 1/8-in. SPHERES

108-5864 Rev.

Figure 53

PECLET NUMBERS FOR
FLUIDIZED-PACKED
BEDS PACKED WITH
3/8 x 3/8-in. CYLINDERS

108-5867 Rev.

In the runs with fluidized material, the peak concentration was not usually at the center of the column, and its position varied with time. The peak concentration of the tracer gas for the fluidized-packed bed usually did not correspond exactly to that of the packed bed of the same type of fixed packing, but it did fall within a factor of 2 or 3. For example, in Table X, data for 12-in. spherical packing indicated that the tracer gas peak concentration for the packed bed ranged from 0.41 to 0.56% carbon dioxide, whereas the peak concentration for the fluidized-packed bed varied from 0.32 to 0.81% carbon dioxide. For 1/8-in. spherical packing (see Table III), the peak concentration for the packed bed ranged from 1.31 to 1.75% carbon dioxide, whereas the peak concentration for the fluidized-packed bed ranged from 0.55 to 1.35% carbon dioxide. This resulted in Peclet numbers that had a greater variance than the packed-bed case and that could not be well-represented by a continuous curve. However, Figures 50, 51, 52 and 53 indicate an agreement within a factor of 2 or 3 between the results of the packed-fluidized and the packed-bed experiments. No definite trend in the relationship between the Peclet number and the Reynolds number with respect to the size or type of fluidizing material was noted.

These results may be referred to a model of fluidized beds presented by Rowe,⁽⁴³⁾ which holds that gas flow patterns determine the solids mixing behavior. In Rowe's analysis, the gas flow is basically uniform and streamlined through the dense particle phase, but the gas flow is diverted by the pressure gradients in the rising bubbles. For the fluidized-packed bed, the gas stream paths are deflected by the fixed packing to give radial displacements of a random-walk type, as in the usual packed bed. But these displacements are also subject to variances caused by rising bubbles. Thus, the resulting eddy diffusivities are apparently determined by the fixed packing, but show larger variances due to the contributions of the motion of the bubbles in the fluidized bed.

For design purposes, the gas mixing for a fluidized-packed-bed reactor can be estimated from packed-bed data with a suitable safety factor to allow for the directional variance in the gas stream caused by nonuniform pressure gradients associated with fluidization.

3. Fluidized Bed without Fixed Packing

For comparison, two runs were made in a fluidized bed without fixed packing, and one run was made with an empty column. The Peclet numbers and Reynolds numbers were based on the column diameter, D_t . Peclet number as a function of Reynolds number for an empty column is shown in Figure 54 and compared with Peclet numbers for the same tube containing 0.0110- and 0.0080-in.-diam glass beads. With gas passing through the bed of 0.0110-in. glass beads at velocity 0.3 ft/sec below the minimum fluidization velocity, the peak concentration was 5.83% carbon dioxide. At a fluidizing gas velocity of 0.4 ft/sec, the peak concentration

fell to 0.56% carbon dioxide. This result indicates essentially laminar flow through the static bed, which thus shows a higher Peclet number than

does the empty column (no packing). This result is consistent with the results of Wace and Burnett,⁽⁵³⁾ who demonstrated essentially streamlined gas flow in fluidized beds near incipient fluidization (gas velocity 1 to 1.25 times minimum fluidization velocity). However, with increasing gas velocity, there is an increase in the turbulent eddies in open-column fluidization until essentially complete gas mixing is reached.

Figure 54
PECLET NUMBERS FOR FLUIDIZED BED
AND FOR AN EMPTY COLUMN

108-5865

turbulence is reduced by the addition of 1/4-in. spherical packing. Fluidizing 0.0110-in. glass beads in the voids of the packing results in a wider scatter of Peclet numbers about the curve for the packed bed, but the relationship between Peclet numbers and Reynolds numbers could be represented approximately by this same curve. The addition of 0.0110-in. glass beads to the column without the fixed packing results in a considerable reduction of the turbulence of the gas at gas velocities below the minimum fluidization velocity. However, as the gas velocity is increased, the turbulence increases until it exceeds that of the tests with the empty column.

F. Conclusions

Turbulent gas mixing (eddy diffusion) in a fluidized-packed bed was essentially the same as in packed beds with the same size fixed packing. However, because of the non-uniform pressure gradients associated with fluidization, there was a larger variance in the eddy diffusivities, and sometimes an asymmetrical concentration profile was observed for the tracer gas with respect to the column.

The addition of nonfluidized particles to a column reduced the turbulent eddies in the gas flow through the tube. However, if the particles were fluidized, eddy mixing increased with increasing gas flow and exceeded that of the open tube.

Figure 55 compares the eddy diffusion for the four different types of systems investigated. Using the empty column as reference, the tur-

Figure 55
COMPARISON OF GAS TURBULENCE
IN VARIOUS SYSTEMS

108-5866

VI. ELUTRIATION

Of major concern in fluidized-bed reactors are the bed losses by elutriation. In fluidized-packed beds, the solids movement is inhibited by the presence of the packing. Elutriation rates should therefore be less in fluidized-packed beds than in fluidized beds. Rates were compared for fluidized-packed beds, fluidized beds, and beds consisting of a lower zone that was fluidized-packed and an upper fluidized zone. So that the elutriation data would have direct application to the pilot-plant development of the fluid-bed fluoride volatility processes, uranosic oxide (U_3O_8) fines were elutriated from beds of alumina grain and 1/2-in., cylindrical, uranium dioxide pellets.

A. Equipment and Experimental Conditions

All runs were conducted in 3-in.-diameter glass columns 3, 5, and 6 ft long. The variation in column length permitted some investigation of the effect of freeboard in the elutriation rates. The bed support and gas distributor consisted of a perforated metal disk, which was either 3 or 1 in. above the flanged interface between the bottom of the column and the conical gas inlet. For the runs using the 6-ft column, there was a 3-in. layer of 1/4-in. steel balls above the distributor plate to improve gas dispersion. The fluidized bed runs were only made in the 6-ft column.

A separate filter chamber was in the off-gas line from the column. The filter chamber could be disconnected and weighed to determine the rate of U_3O_8 fines collection. Humidified air was the fluidizing gas. Because a pulsed gas flow is effective in improving solids motion, a timer and solenoid valve system was installed in the gas inlet to cause a pulsed gas flow to be superimposed upon a steady flow of fluidizing gas. Both the fluidizing gas and the pulsing gas were air.

The packed bed consisted of 1/2-in.-diameter by 1/2-in.-long uranium dioxide pellets. Two sizes of uranosic oxide fines were elutriated having average diameters of 4 and 12 microns. The fines size analysis was made by the Coulter Counter* technique. The average diameter was calculated on a weight-percent basis. Various sizes of fused alumina grain were used as the fluidized particles: -54 +200 mesh, -40 +170 mesh, -40 +80 mesh, -60 +120 mesh, and -60 +170 mesh.

B. Elutriation Studies in the 3- and 5-ft Columns

The use of two column heights was essentially to test the effect of freeboard on the elutriation rates. For the runs with the 3-ft column, the bed heights were adjusted so that the freeboard was about 2 ft, while for the

*Product of Coulter Electronics, Inc., Chicago, Illinois.

runs with the 5-ft column, the bed heights were such that the freeboard was about 3 ft. The conditions for these runs are given in Table XVI. All tests in these columns used 12-micron, U_3O_8 fines and -54 +200 mesh alumina particles and the 1/2-in., cylindrical, uranium dioxide pellets.

Table XVI
CONDITIONS FOR ELUTRIATION TESTS WITH THE 5- AND 3-ft COLUMNS

Elutriation Run	Column Height, ft	Static Height of Alumina Bed Relative to Top of UO_2 Pellet Bed, in.	Air Velocity, ft/sec	Fines Charge, g	Fines Elutriated, g	Support-plate Height above Flange Interface, in.	Bed UO_2 Height, in.	Freeboard at Static Conditions, in.
1	3	+6	0.67	137.3	106.2	3	6	21
2	3	0	0.67	128.4	123.1	3	6	27
3	3	+6	0.67	107.0	98.0	3	6	21
4	3	+6	0.34	115.0	58.3	3	6	21
5	3	0	0.34	197.5	88.1	3	6	27
6	5	+6	0.67	123.0	107.4	1	18	35
7	5	0	0.34	138.6	56.8	1	18	41
8	5	0	0.67	190.3	171.5	1	18	41
9	5	+6	0.34	253.7	134.0	1	18	35
10	5	-9	0.34	190.0	36.1	1	18	41
11	5	-9	0.67	187.0	186.0	1	18	41

The quantity of fines elutriated as a function of time for the various run conditions is shown in Figures 56 and 57. The data indicate that the height of the alumina grain bed height had little effect on the rate of fines

Figure 56

ELUTRIATION RATE FOR 3-ft COLUMN

Figure 57
ELUTRIATION RATE
FOR 5-ft COLUMN

carryover. More significant were the gas velocity and freeboard. In the first 10 to 15 min, the rate of elutriation in the 3-ft column was about twice that in the 5-ft column. A superficial air velocity of 0.67 ft/sec would remove about 90% of the fines charged in 45 min of operation, whereas the velocity of 0.34 ft/sec removed about 50% of the fines charged after the same operating time.

To characterize the rate of elutriation by a parameter or rate constant, Leva⁽²⁶⁾ applied a first-order rate law to elutriation:

$$\frac{dC}{d\theta} = -KC, \quad (48)$$

where

C = Fines concentration in bed,

θ = Time,

and

K = Rate constant.

Assuming K does not vary with C , Equation 48 can be integrated between times 0 and θ to yield

$$\ln(C/C_0) = -K\theta, \quad (49)$$

where

C_0 = Initial fines concentration.

The data for Runs 6 and 9 in terms of $\ln(C/C_0)$ versus time are plotted in Figure 58. The elutriation curves are of the usual shape for a plot of this type in that a first-order rate constant can be approximated from the initial slope of the curve, but the curve deviates rapidly from a straight line. This is evidently because of the assumption that rate constant is independent of fines concentration. Therefore, this simple assumption has limited utility for describing actual data (see Zenz and Othmer.⁽⁵⁹⁾)

In studies on drag forces in a hydraulic model of a fluidized bed, Rowe and Henwood⁽⁴⁵⁾ have shown that the drag on a sphere in a linear chain of spheres is less than the drag on an isolated sphere. The spheres in a chain are shielded in the turbulent wake of the sphere before it. Figure 59 shows the effect of the number of spheres in the chain on the drag ratio between a sphere in the chain and an isolated sphere.

Figure 58

CORRELATION OF ELUTRIATION DATA FOR A RATE CONSTANT INDEPENDENT OF FINES CONCENTRATION (LEVA'S MODEL)

Figure 59

EFFECT OF CHAIN LENGTH ON AVERAGE DRAG PER SPHERE

It can, therefore, be considered that a similar shielding effect might occur in a bed containing relatively large fluidized particles and fines. With

large concentrations of fines, there should be more fine particles exposed to regions of high drag forces. As the fines concentration decreases, the fraction of fines shielded by larger particles would increase. This effect was estimated by a parabola as shown in Figure 60. The drag was assumed to have the following relationship:

$$D_f = K' \sqrt{C}, \quad (50)$$

where

$$K' = \text{a constant,}$$

and

$$D_f = \text{drag.}$$

Figure 60
ASSUMED EFFECT OF
FINES CONCENTRATION
IN A FLUIDIZED BED ON
THE AVERAGE DROP
PER FINE PARTICLE

Applying this concept to a rate equation, where the rate depends upon the concentration to the first power and the rate constant depends on the concentration to the one-half power, yields

$$\frac{dC}{d\theta} = (-K' \sqrt{C}) C, \quad (51)$$

$$\int_{C_0}^C \frac{dC}{C^{3/2}} = \int_0^\theta -K' d\theta, \quad (52)$$

and by integrating

$$\frac{1}{\sqrt{C/C_0}} = K\theta + 1, \quad (53)$$

where $K = (K'/2) \sqrt{C_0}$.

Therefore, a plot of $1/\sqrt{C/C_0}$ versus θ should have a slope of K and an intercept of 1. This is the case as shown in Figure 61, where the data for Runs 6 and 9 are plotted again. The data follow a linear curve for times extending up to 160 min, whereas a plot of $\ln(C/C_0)$ versus time gave a linear fit for a maximum time of 40 min.

Figure 61

CORRELATION OF ELUTRIATION DATA FOR AN
ELUTRIATION RATE CONSTANT DEPENDING UPON
CONCENTRATION TO THE ONE-HALF POWER

Figure 62

CORRELATION OF ELUTRIATION
DATA FOR AN ELUTRIATION
RATE CONSTANT DEPENDING
UPON CONCENTRATION
TO THE FIRST POWER

A plot was also made of $1/(C/C_0)$ versus time, θ , as shown in Figure 62 to see if a good data fit could be obtained if it were assumed that the rate constant was directly proportional to the concentration. Except for the one point at 80 min for the run with the 0.67-ft/sec gas velocity, the data follow a linear plot quite well. The elutriation rate constant depends upon the concentration of fines and is fairly insensitive to the choice of first- or half-power dependency. Thus, by a simple assumption, a rate constant can be determined that describes the elutriation rate over a much greater range than that of the function proposed earlier by Leva.

An extensive study was not made, however, so that the rate constants developed here could not be

correlated for various materials over a wide range of conditions. Experimentation was mainly confined to mock-ups of the pilot plant.

C. Elutriation Studies in the 6-ft Column

The elutriation studies were performed in the 6-ft column, mainly to compare the elutriation rates for fluidized and fluidized-packed beds and to determine the effect of packed-gas flow. All runs in the 6-ft column used a standard mixture of 3100 g of fused alumina grain and 200 g of uranositic oxide fines (average diameter 4 microns). Except for runs made to determine the effect of the alumina grain size on elutriation, -40 +170 mesh particles were used for fluidized material. The alumina grain bed height (static) for all runs was about 18 in., and the initial column freeboard (static conditions) was 54 in. The fluidizing gas was distributed through a perforated brass disk supporting a 3-in. layer of 1/4-in. steel balls. Humidified air was the gas used for fluidizing and pulsing.

The first series of runs in the 6-ft column was made with a fluidized bed (no UO_2 pellet bed). The fluidizing gas velocities chosen for these runs were in the range used in the fluid-bed fluoride volatility processes. In Figure 63, an increase in the elutriation of the U_3O_8 fines is shown for increasing fluidization gas velocity (from 1/3 to 1 ft/sec). The effect of gas pulsation on elutriation is shown in Figure 64. In these experiments, the gas pulse manifold pressure was 10 psig, and the fluidizing gas velocity was 0.67 ft/sec. The duration of the gas pulse was kept constant at 0.3 sec. The elutriation rate of the U_3O_8 fines doubled as the gas pulse rate was

Figure 63

ELUTRIATION OF U_3O_8 FINES FROM A MIXTURE OF U_3O_8 FINES IN FLUIDIZED ALUMINA, AS A FUNCTION OF FLUIDIZING GAS VELOCITY

Equipment:	6-ft-long, 3-in.-diam, glass column
Bed material:	200 g of U_3O_8 (~4-micron diam)
	3100 g of alumina grain (-40 +170 mesh)
Bed height (static):	18 in.
Fluidizing gas:	Air

108-6534

Figure 64

ELUTRIATION OF U_3O_8 FINES FROM
A MIXTURE OF U_3O_8 FINES IN A
FLUIDIZED BED OF ALUMINA, AS A
FUNCTION OF PULSE FREQUENCY

Equipment: 6-ft-long, 3-in.-diam,
glass column
Bed material: 200 g of U_3O_8 fines
(~4-micron diam)
3100 g of alumina grain
(-40 +170 mesh)
Fluidizing and
pulsing gas: Air
Gas pulse manifold
pressure: 10 psig
Superficial gas
velocity: 0.67 ft/sec
Pulse duration: 0.3 sec

Figure 65

ELUTRIATION OF U_3O_8 FINES FROM A
MIXTURE OF U_3O_8 FINES IN A FLUIDIZED
BED OF ALUMINA, AS A FUNCTION OF
PULSE GAS MANIFOLD PRESSURE

Equipment: 6-ft-long, 3-in.-diam,
glass column
Bed material: 200 g of U_3O_8 fines
(~4-micron diam)
3100 g of alumina grain
(-40 +170 mesh)
Fluidizing and
pulsing gas: Air
Pulse duration: 0.3 sec
Superficial fluidizing
gas velocity: 0.67 ft/sec
Gas pulse frequency: 10 pulses/min

108-6535

increased from 0 to 20 pulses/min. Figure 65 shows an increase in elutriation rate for increasing gas manifold pressure (from 10 to 30 psig) at a constant gas pulse frequency of 10 pulses of 0.3-sec duration per minute.

The elutriation rate of U_3O_8 fines from a fluidized-packed bed was then studied as a function of the uranium dioxide pellet bed height, pulse-gas frequency, pulse-gas manifold pressure, and alumina grain particle size. Uranium dioxide pellets, 1/2 in. in diameter by 1/2 in. long, were used for the packing, and the fluidized material was composed of U_3O_8 fines (4-micron diameter) and the -40 +170 mesh alumina grain. Air, at a superficial velocity of 2/3 ft/sec, was used as the fluidizing gas. The results of these studies are shown in Figures 66, 67, and 68.

Figure 66

ELUTRIATION OF U_3O_8 FINES FROM A MIXTURE OF U_3O_8 FINES IN FLUIDIZED ALUMINA, AS A FUNCTION OF UO_2 PELLET BED HEIGHT AND GAS PULSE FREQUENCY

Equipment: 6-ft-long, 3-in.-diam, glass column
 Bed material: 200 g of U_3O_8 fines (~4-micron diam)
 3100 g of alumina grain (-40 +170 mesh)
 Fluidizing and pulsing
 gas velocity: 0.67 ft/sec
 Gas pulse pressure: 10 psig
 Pulse duration: 0.3 sec

108-6536

Figure 67

ELUTRIATION OF U_3O_8 FINES FROM A MIXTURE OF U_3O_8 FINES IN FLUIDIZED ALUMINA, AS A FUNCTION OF PULSE GAS MANIFOLD PRESSURE AND UO_2 PELLET BED HEIGHT

Equipment: 6-ft-long, 3-in.-diam, glass column
 Bed material: 200 g of U_3O_8 fines (~4-micron diam)
 3100 g of alumina grain (-40 +170 mesh)
 Fluidizing and pulsing gas: Air
 Superficial fluidizing gas velocity: 0.67 ft/sec
 Gas pulse frequency: 10 pulses/min
 Pulse duration: 0.3 sec

108-6537

Figure 68

ELUTRIATION OF U_3O_8 FINES FROM A MIXTURE OF U_3O_8 FINES IN FLUIDIZED ALUMINA IN A PACKED BED, AS A FUNCTION OF ALUMINA PARTICLE SIZE

Equipment: 6-ft-long, 3-in.-diam, glass column
 UO_2 pellet bed height: 9 in.
 Bed material: 200 g of U_3O_8 fines (~4-micron diam)
 3100 g of alumina grain
 Fluidizing gas: Air
 Superficial fluidizing gas velocity: 0.67 ft/sec
 No gas pulsing

108-6538

Figures 66 and 67 show that the elutriation rates were generally lower in those runs in which the fluidized bed was packed with a bed of uranium dioxide pellets than in the runs in which the fluidization was unhindered by the presence of the packing. In the two runs made with 18-in.-deep beds of uranium dioxide pellets, the composition of the fluidized bed was the same as that used in runs with 9-in.-deep pellet beds, but the amount of the U_3O_8 -alumina mixture was adjusted so that the fluidized-bed portion did not extend beyond the top of the pellet bed. For these runs (18-in.-deep pellet beds), 60 g of U_3O_8 fines and 760 g of alumina grain were charged to the column. In one of these runs, gas pulsing was not used. The rate of elutriation for the run without gas pulsing was very low. The uranium dioxide pellets inhibited the transport of the U_3O_8 fines. However, gas pulsing overcame this effect.

In the runs that investigated the elutriation rate from a fluidized-packed bed as a function of the size of the alumina grain, the elutriation rates increased with decreased grain size (Figure 68). However, up until 85% of the fines in the No. 90 (90-120 mesh) alumina grain were elutriated, maximum elutriation rates were obtained with the No. 90 alumina grain. The lower elutriation rates for the larger fluidized particles may be because of lower particle mixing. With the smallest alumina particles (No. 60), increased particle cohesion may have caused a decreased rate of fines elutriation.

D. Conclusions

Although these studies did not lend themselves to a generalized correlation for elutriation rates, the following conclusions can be drawn:

- 1) Elutriation rates are a strong function of gas velocity. Rates increase by a factor of about 10 as the gas velocity increases from 0.33 to 1.0 ft/sec.
- 2) Bed height does not seem to have a significant effect on elutriation. However, of more significance is the freeboard. By increasing the column height from 3 to 5 ft (2-ft increase in freeboard) elutriation rates were halved.
- 3) Elutriation rates increase with both pulse frequency and pressure.
- 4) Elutriation is hindered by the presence of a fixed uranium dioxide pellet bed.
- 5) Maximum elutriation rates of 4-micron U_3O_8 fines were attained from a fluidized bed of alumina grain that was about 90-120 mesh in size.
- 6) Rates of elutriation are best interpreted if the rate constant is assumed to be a function of the concentration.

VII. CONCLUSIONS

The results of the experimental and theoretical studies can be summarized as follows:

1) Fluidization for a fluidized-packed bed was quite smooth, and slugging did not occur at superficial gas velocities that would cause violent slugging in a fluidized bed.

2) Lateral solids mixing in a fluidized-packed bed can be described by

$$D = 0.22(0.0558 D_p) \left[\frac{V - V_{mf}}{V_{mf}} \right] (V_{mf}^{0.0054}) \left[\frac{\epsilon_B^{0.0054}}{\epsilon_B} \right].$$

3) A correlation for longitudinal solids mixing was not developed; but, in general, mixing rates in the longitudinal direction were about an order of magnitude greater than those in the lateral direction.

4) Highest rates of longitudinal solids mixing were obtained with the -100 +120 mesh copper-nickel shot than for any other size of fluidized copper-nickel shot studied.

5) Longitudinal mixing of solids was about an order of magnitude less for a fluidized-packed bed than for normal fluidization without the presence of packing.

6) The diffusion coefficient for the upward transport of 24-micron nickel fines through a fluidized-packed bed, consisting of 3/8-in. spherical packing and -40 +170 mesh alumina grain with a superficial fluidizing gas velocity of 0.8 ft/sec, was about 10^{-2} ft²/sec.

7) The lateral thermal conductivity of a fluidized-packed bed can be correlated as follows:

$$\frac{k_e}{\rho C_p} = (0.0075) 0.0558 D_p \left(\frac{V - V_{mf}}{V_{mf}} \right).$$

8) A correlation for longitudinal thermal conductivity of a fluidized-packed bed was not developed, but values for longitudinal thermal conductivities were more than 20 higher than those for lateral thermal conductivities.

9) Heat-transfer coefficients at the column wall are up to a factor of two less in a fluidized-packed bed than for the same fluidized particle without packing.

10) The heat-transfer coefficient of the packing surface is increased by a factor of about eight with the addition of fluidized particles in the voids of the fixed packing.

11) Eddy diffusion in a fluidized-packed bed was essentially the same as in packed beds with the same size fixed packing. However, because of the nonuniform pressure gradients associated with fluidization, there was a larger variance in the eddy diffusivities for the fluidized-packed bed.

12) Elutriation of fines is decreased by the addition of fixed packing of a fluidized bed.

VIII. DESIGN APPLICATIONS

A. Direct Fluorination

Initial pilot-plant development for the recovery of fissionable materials from uranium dioxide pellet-type fuels was by a direct fluorination process. Fluorine reacted with the uranium dioxide pellets to form the volatile uranium hexafluoride according to this overall reaction:

A flow schematic diagram for the direct fluorination process is shown in Figure 69. Because of the highly exothermic nature of this reaction,

Figure 69

FLOW-SCHEMATIC DIAGRAM FOR
DIRECT FLUORINATION OF UO_2

an inert material (either magnesium fluoride, calcium fluoride, or alumina grain) was fluidized in the voids of the reacting uranium dioxide pellets. External fins were provided for forced-air cooling, and the column was heated by electrical resistance heaters.

The reactant gas consisted of fluorine and oxygen with nitrogen diluent. The oxygen occurred as a by-product from the fluorination of the uranium dioxide (see Figure 69). The off-gas, consisting of oxygen, nitrogen, and unreacted fluorine from the con-

denser, either went entirely out into the scrubber or was recycled. For the recycling, the oxygen concentration increased in the reactant gas and was greater than if the off-gas was released directly from the scrubber. The oxygen increased the UF_6 production rate by reacting with the uranium dioxide pellets to form uranosic oxide (U_3O_8), which was in the form of fines. The high surface area of the fines caused an increased rate of reaction.

1. Heat Transfer

Temperature drops across the reacting bed and at the wall during direct fluorination processing can be calculated from the results of the heat transfer studies for fluidized-packed beds.

As an example, the following processing conditions will be assumed:

3-in.-diameter reactor,
 6-in.-deep UO_2 pellet bed,
 -40 +200 mesh alumina grain (average diameter 0.2 mm),
 Reaction temperature = 500°C ,
 UF_6 production rate = 0.7 kg/hr,
 Gas flow = 0.71 cfm (25°C) or 0.63 ft/sec (500°C),
 Fixed bed voidage, $\epsilon = 0.4$, and
 Average pellet size equivalent to a 3/8-in.-diameter sphere.

2. Temperature Drop across Bed and Wall

First the minimum fluidization velocity for the 0.2 mm alumina grain is determined. The method of Rowe⁽⁴⁴⁾ can be used. Rowe's method is described by a nomograph in the appendix for convenience in calculation. The following data are necessary for determination of the minimum fluidization velocity:

$$\begin{aligned}d &= 0.2 \text{ mm}, \\ \rho_s &= 4 \text{ g/cm}^3 \text{ or } 249 \text{ lb/ft}^3, \\ \mu &= 0.036 \text{ centipoise (N}_2 \text{ at } 500^\circ\text{C)}, \text{ and} \\ \rho_g &= 0.0044 \text{ g/cm}^3 \text{ (N}_2 \text{ at } 500^\circ\text{C)}.\end{aligned}$$

It is assumed that the F_2 and O_2 concentrations are low and the properties of the reactor gas can be represented by N_2 . From the nomograph, the minimum fluidization velocity, V_{mf} , is estimated to be 0.34 ft/sec. The maximum allowable gas velocity or terminal velocity of the alumina particles can also be quickly determined from the nomograph and is found to be 8.4 ft/sec.

Figure 35 (Sec. IV-A) gives the general correlation for lateral thermal diffusivities in a fluidized-packed bed as a function of $0.0558 D_p(V - V_{mf}/V_{mf})$.

$$\begin{aligned}0.0558 D_p(V - V_{mf}/V_{mf}) &= 0.0558 \left(\frac{3}{8 \times 12} \right) \left[\frac{\frac{0.63}{0.4} - 0.34}{0.34} \right] \\ &= 6.3 \times 10^{-3}\end{aligned}$$

From Figure 35:

$$\begin{aligned}\frac{k_e}{\rho C_p} &= \alpha' = 5 \times 10^{-5} \text{ ft}^2/\text{sec} \\ k_e &= (5 \times 10^{-5})(4 \times 62.4)(0.217)(3600) \\ &= 9.8 \text{ (Btu/hr)(ft}^2\text{)(}^\circ\text{F/ft)}.\end{aligned}$$

However, the correlation on Figure 35 derived from heat-transfer measurements at 50°C. The theoretical discussion in Sec. IV-A indicates that the effective bed thermal conductivity would be proportional to the gas thermal conductivity. Since the process gas temperature would be about 500°C, the higher thermal conductivity of the gas should be considered. Therefore

$$\text{at } 50^{\circ}\text{C, } k_g = 0.0162;$$

$$\text{at } 500^{\circ}\text{C, } k_g = 0.0325;$$

$$9.8(0.0325/0.0162) = 20.$$

In any case, the thermal diffusivity cannot exceed the solids mixing diffusivity (refer to Sec. IV-A-1 for discussion on lateral heat transfer). For these conditions, the correlation on Figure 18 (Sec. III-A) gives a D of $4 \times 10^{-4} \text{ ft}^2/\text{sec}$.

From Table IV, the density of the alumina grain bed at a V of 0.63/0.4 (or 1.58) ft/sec is estimated to 42 lb/ft³. Then,

$$k_e = (4 \times 10^{-4})(42)(0.217)(3600) = 13 \text{ Btu}/(\text{hr})(\text{ft}^2)(^{\circ}\text{F}/\text{ft}).$$

Since the thermal conductivity of the bed is limited by the heat content of the moving solids, the thermal conductivity determined on the basis of solids mixing will have to be used for the elevated temperatures.

Because of insufficient data, a generalized correlation for wall heat-transfer coefficients was not developed. Therefore, it is assumed that data obtained for glass beads (Figure 41) is sufficiently close to that expected for alumina grain. Equation 28 shows that the increase in gas thermal conductivity must be compensated for.

Table XVII indicates the thermal conductivities and wall heat-transfer coefficients for various gas velocities.

Table XVII

HEAT-TRANSFER COEFFICIENTS AND THERMAL CONDUCTIVITIES
FOR FLUIDIZED-PACKED BED OF UO_2 PELLETS AND
0.2-mm ALUMINA GRAIN

Superficial Gas Velocity, V_0 , ft/sec	Gas Velocity Corrected for Packing Voidage, V , ft/sec	Fluidized- bed Density, lb/ft ³	Thermal Conductivity at 500°C, k_e , Btu/(hr)(ft ²) (°F/ft)	Heat-transfer Coefficient at 500°C, h , Btu/(hr)(ft ²) (°F)
0.44	1.10	45	9	53
0.63	1.58	42	13	70
0.88	2.20	36	18	97
1.10	2.75	31	19	114

The following assumptions are made in estimating the temperature drops from the center of the bed to the column wall:

- 1) Heat transfer to the cooling wall is entirely in the radial direction.
- 2) Heat generation is uniform in the pellet-bed zone.

Then,

$$\Delta t_{\text{bed}} = \frac{q'}{k_e} \left(\frac{r_b^2}{4} \right), \quad (55)$$

where

Δt_{bed} = Temperature difference across radius;

q' = Rate of heat generation, Btu/(hr)(ft³);

k_e = Lateral thermal conductivity, Btu/(hr)(ft²)(°F/ft);

and

r_b = Radius of bed, ft.

Thus,

$$\Delta t_{\text{bed}} = \frac{(700/352)(259)(1728)}{\pi(1.5)^2(6)(13)(0.252)} \times \frac{(1.5)^2}{4} \times \frac{1}{144} \times \frac{1}{1.8} = 14^\circ\text{C}.$$

The temperature drop at the wall would be

$$\Delta t_{\text{wall}} = \frac{q}{hA}. \quad (56)$$

Usually the charge of alumina grain is such that the height of the alumina particles exceeds the height of the bed of uranium dioxide pellets. Since the vertical movement of the fluidized particles is very rapid, the heat transfer at the wall will be effectively through an area greater than the small area in contact with the uranium dioxide pellet bed. For calculation purposes, it is assumed that this area is effectively twice the area of the wall in contact with the uranium dioxide pellet bed. Thus,

$$A = (2)(6)\pi(3)(1/144) = 0.785 \text{ ft}^2.$$

Therefore,

$$\Delta t_{\text{wall}} = \frac{(700/352)(259)}{(70)(0.785)(0.252)(1.8)} = 21^\circ\text{C}.$$

Table XVIII compares the temperature differentials obtained during pilot-plant fluorinations⁽¹⁷⁾ and those calculated on the basis of the results from heat-transfer tests. The conditions for Run 32 are comparable to those used for this example. In Run 34, the off gas was recycled without any nitrogen addition. The oxygen reacted to form uranous oxide fines, which increased the reaction rate; therefore, the rate of heat generation was increased. Good agreement is shown between the experimental and calculated values.

Table XVIII
COMPARISON OF TEMPERATURE DIFFERENTIALS
OBTAINED DURING FLUORINATION WITH THOSE
CALCULATED FROM HEAT-TRANSFER TESTS

6-in.-deep bed of UO_2 pellets
Control temperature = 500°C

Run Number	32	34
Diluent gas	N_2	O_2
Superficial gas velocity, V_0 , ft/sec	0.63	0.71
UF_6 collection rate, kg/hr	0.70	1.25
Heat generation, Btu/(hr)(ft ³)	83,000	149,000
Measured Δt , $^\circ\text{C}$	30	60
k_e , Btu/(hr)(ft ²)($^\circ\text{F}/\text{ft}$)	13	13*
Calculated Δt_{bed} , $^\circ\text{C}$	14	25
h , Btu/(hr)(ft ²)($^\circ\text{F}$)	70	70*
Calculated Δt_{wall} , $^\circ\text{C}$	21	37
Total calculated Δt , $^\circ\text{C}$	34	62

* k_e and h are for a superficial velocity of 0.63 ft/sec. The effect of the slightly increased gas velocity (0.71 ft/sec) was neglected.

Figure 70 predicts the total temperature differential from the center of the bed to the wall for various uranium hexafluoride production rates and superficial fluidizing gas velocities. The data from the two pilot-plant runs are also indicated on Figure 70.

Figure 70
TEMPERATURE DIFFERENTIAL
BETWEEN CENTER OF BED AND
WALL FOR DIRECT FLUORINATION
OF UO_2 PELLETS

3. Gas Mixing and Elutriation

Since the entire reactant gas enters at the bottom of the column, the gas would be completely mixed, and the fluorine can be considered evenly distributed throughout the bed.

Generally, conditions are adjusted in the direct fluorination process to keep fines production at a minimum. Also, the filters are blown back at regular intervals to return elutriated fines to the reaction zone. Therefore, fines elutriation are not of principal interest in the direct fluorination process. The subject of fines elutriation will be considered in the two-zone fluorination process described immediately below.

B. Two-zone Fluorination

In the previously described direct fluorination process, one limitation was the depth of the uranium dioxide pellet bed. If the bed were too deep, the uranium hexafluoride formed in the lower sections of the bed would react with the uranium dioxide in the upper portion to produce intermediate uranium fluorides, which would form in layers on the pellet surfaces and cause caking. Therefore, pilot-plant efforts were directed to a two-zone fluorination scheme. In the two-zone fluorination scheme, oxygen and nitrogen are introduced at the bottom of the column. The oxygen reacts with the uranium dioxide pellets to form uranosic oxide fines, which are then carried up in the inert fluidized bed above the bed of uranium dioxide pellets. Fluorine is injected just above the pellet bed to fluorinate the fines to uranium hexafluoride. The inert fluidized bed extends about 2 ft above the bed of uranium dioxide pellets.

1. Heat Transfer

Since the heat of reaction for the oxidation of uranium dioxide (19.3 kcal/g-mole) is less than the heat of reaction for the fluorination of uranium dioxide (259 kcal/g-mole), there would be a correspondingly lower rate of heat generation and hence temperature differential in the fluidized-packed-bed portion of the reactor. For example, for conditions comparable to Run 34 of the direct fluorination process with an uranium hexafluoride production rate of 1.25 kg/hr, the temperature drop across the fluidized-packed uranium dioxide pellet bed would be 1.8°C if a two-zone processing scheme would be used. For the direct fluorination process, the temperature drop was calculated to be 24.7°C for the same production rate. In the open-tube fluidized portion of the two-zone reactor, there would be a greater rate of heat generation (about 240 kcal/g-mole) caused by the reaction of the uranosic oxide fines with fluorine. However, in this zone there is free fluidization unhindered by uranium dioxide pellets, and therefore temperature differentials would likewise be low.

From the standpoint of temperature drop across the bed, process control appears very good for the two-zone fluorination scheme. However, because of the high concentration of uranosic oxide fines, the possibility of ignitions and resultant sintering of the fluidized particles is of concern.

The temperature of the particle throughout its life is described by the following equation, which relates heat capacity, generation of chemical heat, and heat losses through conduction and radiation:

$$(4/3) \pi r^3 \rho_s C_p \frac{dT}{d\theta} = K(4\pi r^2) \Delta H - 4\pi r^2 h(T - T_s) - 4\pi r^2 \sigma \epsilon_h (T^4 - T_s^4), \quad (57)$$

where

r = Radial coordinate,

ρ_s = Density of particle,

C_p = Heat capacity,

K = Rate of reaction,

ΔH = Heat of reaction,

T = Temperature,

θ = Time,

T_s = Temperature of surroundings,

σ = Stefan-Boltzmann constant,

and

ϵ_h = Emissivity.

If, for simplicity, the heat capacity of the particle is neglected, the above equation reduces to

$$K\Delta H = h(T - T_s) + \epsilon_h \sigma (T^4 - T_s^4). \quad (58)$$

The rate of heat transfer will depend upon the rate of reaction. A determination must be made to show whether the rate is controlled by fluorine diffusion through the gas film or by chemical kinetics.

From the Hirschfelder equation,⁽⁴¹⁾ the diffusivity of fluorine in nitrogen at 500°C is estimated to be 0.95 cm²/sec. Because of the small diameter of fine particles (average diameter approximately 7 microns), the Reynolds number for these particles would be very low. Therefore it is assumed that the mass transfer coefficient can be estimated as the minimum value for a sphere in a static medium. In this case,

$$\frac{k_c d}{D_v} = 2,$$

or

$$k_c = \frac{2 \times 0.95}{7 \times 10^{-4}} = 2.72 \times 10^3 \text{ cm/sec},$$

where

k_c = Mass transfer coefficient,

d = Particle diameter,

and

D_v = Molecular diffusivity.

By definition,

$$Na = \frac{k_c}{RT} (p_1 - p_2),$$

where

p = Partial pressure,

and

Na = Rate of mass transfer.

Then,

$$\begin{aligned} \frac{k_c}{RT} &= \frac{2.72 \times 10^3}{82.1 \times 773} = 0.0428 \text{ g-mole}/(\text{cm}^2)(\text{atm})(\text{sec}) \\ &= 154 \text{ g-mole}/(\text{cm}^2)(\text{atm})(\text{hr}). \end{aligned}$$

Using the rate of fluorination of UO_3 powder from boat experiments as an estimate of U_3O_8 fluorination rates at 500°C , the rate of fluorination would be $2.0 \text{ g U}/(\text{cm}^2)(\text{hr})$, with an activation energy of 14.3 kcal/g-mole . The reaction rate coefficient, k_r , would then be

$$\frac{2 \times 3}{238} = 0.0252 \text{ g-mole}/(\text{cm}^2)(\text{atm})(\text{hr}).$$

Considering mass transfer through the film and reaction at the particle surface as series processes, the rate of fluorine depletion is

$$\frac{1}{(1/k_c) + (1/k_r)} (p_1 - p_2) = 0.0252 (p_1 - p_2) \text{ g-mole}/(\text{cm}^2)(\text{hr}). \quad (59)$$

It will be assumed, for demonstration purposes, that p_2 equals zero (fluorine reacts completely at particle surface). It is seen from Equation 59 that essentially all the resistance is in the reaction at the particle surface, and therefore the rate of reaction is controlled by the chemical kinetics.

The heat of reaction per atom of uranium for the fluorination of uranosic oxide is 240 kcal. As for the diffusion case, it will be assumed that the fine particle is spherical. It will also be assumed that the Reynolds number is low, and therefore

$$\frac{hd}{k_g} = 2, \quad (60)$$

where

h = Heat-transfer coefficient,

and

k_g = Thermal conductivity of gas.

The estimated h for the conditions under consideration is

$$\begin{aligned} h &= \frac{0.475 \times 2}{7 \times 10^{-4}} = 1360 \text{ cal}/(\text{hr})(\text{cm}^2)(^\circ\text{C}) \\ &= 2780 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F}). \end{aligned}$$

The emissivity for the particle will be assumed to be 0.8. The equation for determining the particle temperature for 10% fluorine in nitrogen as the reaction gas can now be resolved to

$$\begin{aligned} (0.1)(0.0252)(240,000) &= 1360 (T - 500) + 4.88 \times 10^{-9} (0.8)[(T + 273)^4 - 773^4]; \\ 612 &= 1360 (T - 500) + 3.9 \times 10^{-9} [(T + 273)^4 - 773^4]; \\ T &= 500.4^\circ\text{C}. \end{aligned}$$

This indicates that because of the high heat transfer by film conduction, the particle temperature is essentially that of the bed. However, this calculation is for a single particle of very small diameter. If there is particle agglomeration and the effective diameter of the uranosic oxide fines increases, there would be a corresponding decrease in the heat-transfer coefficient (on the basis of a constant Nusselt number of 2). However, some increase in Nusselt number with agglomerate diameter

would be expected, because of the increase in Reynold's number. The following table shows the relation between effective agglomerate diameter, d , and the film heat-transfer coefficient, h .

d	h , Btu/(hr)(ft ²)(°F)
7 microns	2780
14 microns	1890
21 microns	927
50 microns	389
100 microns	195
870 microns	22.4
0.1 in.	7.7
0.5 in.	1.5
1.0 in.	0.8

On the basis of this analysis, an agglomerate with an effective diameter of 870 microns (0.034 in.) would have a temperature of 700°C. At 700°C, the reaction rate is 13.5 g of U/cm² hr. An agglomerate with a larger diameter (about 0.05 in.) would ignite. It would then appear that bed caking would occur, not by the increased rate of reaction associated with the increased surface area of fines, but by poor distribution and separation of the fines in the fluidized bed.

2. Fluorine Injector Design

Since the fluorine is not thoroughly mixed with the inlet reactor gas at the bottom of the column, but is introduced at a point higher in the column, the distribution of fluorine in the reactor is an important consideration. The assumed conditions for a single fluorine injector are as follows:

Assumptions: Isothermal conditions throughout entire fluidized bed

Constant reaction rate

Slug flow, i.e., no velocity profile

Velocity in fluorine injector same as main gas stream

Longitudinal diffusion negligible

First-order reaction

Conditions: Large-diameter reactor

0.20 w/o fines per cc of alumina grain

0.0080-in.-diameter fluidized alumina grain

7-micron fines

0.5-ft/sec superficial gas velocity

500°C reactor temperature

0.20-in.-ID injector

At steady-state conditions, the partial differential equation describing the fluorine distribution by eddy mixing, assuming no depletion by chemical reaction, is

$$\frac{\delta C}{\delta z} = \frac{E_t}{V_0} \left[\frac{\delta^2 C}{\delta r^2} + \frac{1}{r} \frac{\delta C}{\delta r} \right], \quad (61)$$

where

z = Longitudinal coordinate, ft;

E_t = Total diffusivity (eddy plus molecular), ft^2/sec ;

V_0 = Velocity, ft/sec ;

C = Concentration of fluorine;

and

r = Radial coordinate, ft.

The solution to this equation for z not close to the source of injection is (59)

$$C = \frac{V_0}{E_t} \frac{A_r^2}{4z} e^{-(V_0/E_t)(r^2/4z)}, \quad (62)$$

where

r = radius of injector tube.

However, with depletion by chemical reaction, the partial differential equation describing the fluorine distribution is

$$\frac{\delta C}{\delta z} = \frac{E_t}{V_0} \left[\frac{\delta^2 C}{\delta r^2} + \frac{1}{r} \frac{\delta C}{\delta r} \right] - \frac{KC}{V_0} \quad (63)$$

where

K = rate of reaction, sec^{-1} .

Let

$$C = \phi e^{-Kz/V_0}. \quad (64)$$

Then,

$$\begin{aligned} \frac{\delta C}{\delta z} &= \frac{\delta \phi}{\delta z} e^{-Kz/V_0} - \frac{K}{V_0} \phi e^{-Kz/V_0}, \\ \frac{\delta \phi}{\delta z} e^{-Kz/V_0} - \frac{K}{V_0} \phi e^{-Kz/V_0} &= \frac{E_t}{V_0} \left[\frac{\delta^2 \phi}{\delta r^2} + \frac{1}{r} \frac{\delta \phi}{\delta r} \right] e^{-Kz/V_0} - \frac{KC}{V_0}; \\ \frac{\delta \phi}{\delta z} &= \frac{E_t}{V_0} \left[\frac{\delta^2 \phi}{\delta r^2} + \frac{1}{r} \frac{\delta \phi}{\delta r} \right]. \end{aligned} \quad (65)$$

The homogeneous boundary conditions on C,

$$r = 0, \frac{dC}{dr} = 0;$$

and

$$r = \infty, C = 0;$$

are also satisfied by ϕ . The condition on C at $z = 0$ is necessarily satisfied by ϕ since at $z = 0$, $C = \phi$. Therefore,

$$C(\text{reaction}) = C(\text{no reaction}) e^{-Kz/V};$$

$$C(\text{reaction}) = \frac{V_0}{E} \frac{A_r^2}{4z} e^{-(V_0/E)(r^2/4z)} e^{-Kz/V}. \quad (66)$$

The rate of reaction for the fluorination of UO_3 at 500°C is 2.0/238 g-moles of UO_3 per $\text{cm}^2\text{-hr}$ for one atmosphere of fluorine.

Density of alumina particles at static conditions = 1.9 g/cc;

Density of U_3O_8 fines = approximately 8 g/cc.

Surface area for a bed of 0.2 w/o fines/cc alumina at static conditions, for fine particles of 7-micron average diameter, is

$$\frac{(0.002)(1.9)}{8} \times \frac{4\pi(3.5)^2(10^{-4})^2}{4/3\pi(3.5)^3(10^{-4})^3} = 4.07 \text{ cm}^2.$$

For an expanded fluidized bed, the voidage is estimated to be 0.5. Therefore, the fines surface area would be 2.04 cm^2 . The rate constant for fluorine consumption is

$$K = (2.04)(3)(2.0/238)(22.4)(773/273)(1000)(1/3600) = 0.9 \text{ sec}^{-1}.$$

From Table XIV, eddy diffusivity plus molecular diffusivity is found to be $16.2 \text{ ft}^2/\text{hr}$.

From these data, $C(\text{reaction})$ can be determined. The concentration profiles at various heights above the fluorine injector are shown in Figure 71 for the case of diffusion only (without reaction) and for diffusion with chemical reaction.

Figure 71
 FLUORINE DISTRIBUTION FOR A
 SINGLE INJECTOR FOR
 TWO-ZONE FLUORINATION

The gas mixing would be very rapid, and for reactors of pilot-plant scale (i.e., not greater than 6-in. in diameter), the fluorine distribution from a single injector tube would be quite satisfactory. The expected fluorine efficiencies at various reactor heights are tabulated in Table XIX.

Table XIX
FLUORINE EFFICIENCIES AT
DIFFERENT HEIGHTS ABOVE
GAS INJECTOR

z, ft	F ₂ Efficiency, %
0.25	36
0.50	59
0.75	74
1.00	83

Also, for the conditions assumed for these calculations, fluorine depletion caused by reaction decreases the fluorine concentration by a constant factor at all points on the concentration profile.

3. Fines Transport

To avoid accumulation of oxide fines in the lower oxidation zone of a two-zone reactor, the oxidation rate should not exceed the fines-transport rate.

The general equation for steady-state fines transport in a fluidized bed is given by

$$N_f = D \frac{(C_0 - C_f)}{z}, \quad (67)$$

where

N_f = Rate of fines transport, lb/(ft²)(sec);

D = Fines-transport diffusivity, ft²/sec;

C_0 = Fines concentration in oxidation zone, lb/ft³;

C_f = Fines concentration in fluorination zone, lb/ft³;

and

z = Transport distance (axial), ft.

Thus, the rate of fines transport, N_f , as calculated from the above equation, should not exceed the rate of fines formation by oxidation. If, for example, the diffusivity of 8×10^{-3} ft/sec (for the transport of nickel fines at an air velocity of 0.8 ft/sec in a bed of 3/8-in.-diameter spherical packing and -40 +170 mesh alumina grain) can be applied to U_3O_8 fines transport during pilot-plant processing, the rate at which the fines would be removed from the oxidation zone to the fluorination zone can be estimated. The diffusivity was measured for 24.3 w/o nickel in alumina grain, or a density of nickel fines of about 32.5 lb/ft³ (corrected for packing voidage). If it is desired that the percent of fines does not exceed 10% (13.4 lb/ft³) in the fluorination zone at a distance one foot away, the required fines transport rate can thus be calculated:

$$N_f = 8 \times 10^{-3} \frac{(32.5 - 13.4)}{1} = 0.15 \text{ lb/ft}^2 \text{ sec.}$$

For a 3-in.-diameter reactor, the maximum rate of fines formation that could be tolerated would then be 27 lb/hr. This is well in excess of current reaction rates of about 2 lb/hr.

If it is desired that the rate of fines transport be at least 3 lb/hr and the fines concentration in the oxidation zone not exceed 10 w/o, the maximum fines concentration in the fluorination zone can be calculated in a similar manner, with the assumption that the diffusivity is independent of the fines concentration. This calculation yields a maximum of 8.5 w/o fines in the fluorination zone, to obtain the specified fines-transport rate. This calculated result appears to indicate also that the longitudinal transport of fines proceeds at a practically high rate.

Because of the limited data, these calculations apply only for a gas velocity of 0.8 ft/sec and 3/8-in.-diameter spherical packing. The effect of change in gas velocity, packing diameter, size of fines, and size of the fluidized particles has not been determined. However, rate of fines transport is expected to exceed the oxidation rates and, therefore, accumulation of fines in the lower uranium dioxide bed should not be a problem. Also, as shown on Figure 31, the fines should be fairly uniformly distributed throughout the bed, which should result in uniformity of heat generation and good temperature control.

4. Particle Elutriation

Because of the high production rates of fines, the question of particle elutriation from the reacting bed must be considered in a two-zone processing scheme. Although fluorination is occurring to eliminate the fines, there is some chance that there will be fines escaping, because the distribution of residence times of fines in the fluidized bed is of a statistical nature.

For superficial gas velocities of 0.34 and 0.67 ft/sec, the elutriation rate constants, K_E , for 12-micron-diameter particles are 3.4×10^{-3} and $22 \times 10^{-3} C_0^{1/2} \text{ min}^{-1}$, respectively (Figure 61). The fines charge (253.7 g) for the measurement at 0.67 ft/sec was about twice that (123 g) used for the measurement at 0.34 ft/sec. Since the rate constant is concentration-dependent, the rate constant at 0.67 ft/sec can be determined for the same initial fines charge as at 0.34 ft/sec by dividing by $\sqrt{2}$. Thus, $22/\sqrt{2} = 1.56 \times 10^{-3} C_0^{1/2} \text{ min}^{-1}$ (for initial charge of 123 g of fines).

The fraction of fines elutriated, E_f , can be determined as a function of time as follows:

$$\frac{dE_f}{d\theta} = 2K_E(E_f - 1)^{3/2}; \quad (68)$$

$$\int_0^{E_f} \frac{dE_f}{(1 - E_f)^{3/2}} = \int_0^\theta 2 K_E d\theta; \quad (69)$$

$$\frac{1}{(1 - E_f)^{1/2}} = K_E \theta + 1. \quad (70)$$

The time, θ_R , for a particle to react completely can be calculated from a diminishing-sphere model.

From boat experiments, the reaction rate is 2 g/cm²-hr for the fluorination of UO₃ at 500°C. Then,

$$-\rho_s \frac{dV_s}{d\theta} = 2.0 (4\pi r^2) \text{ g/hr}, \quad (71)$$

where

V_s = volume of sphere.

Thus,

$$dV = d\left(\frac{4}{3} \pi r^3\right) = 4\pi r^2 dr; \quad (72)$$

$$-4\pi r^2 \rho \frac{dr}{d\theta} = (2.0)(4\pi r^2). \quad (73)$$

If the density of the fines particles is taken to be 11 g/cm³,

$$\theta_R = \int_0^\theta d\theta = \int_0^r \frac{(11)(4\pi r^2) dr}{(2.0)(4\pi r^2)} = 5.5 \int_0^r dr \quad (74)$$

For a 12-micron sphere, and an average fluorine concentration of 10%,

$$\theta_R = 5.5 \left(\frac{12}{2} \times 10^{-4}\right) \left(\frac{1}{0.1}\right) (60) = 2.0 \text{ min.}$$

The following assumptions are made:

- 1) Particle reaction occurs only while the particle is in the fluidized bed.

- 2) Particle elutriation rate does not change with particle size. Then the total fraction of particles, F_f , escaping from the

$$F_f = \int_0^\theta (\text{Particle size})(\text{Number of particles elutriated/ time}) d\theta;$$

$$F_f = \int_0^\theta \frac{(6 - 6\theta/\theta_R)^3}{6^3} (2K_E)(F_f - 1)^{3/2} d\theta \quad (75)$$

$$= \int_0^\theta (1 - \theta/\theta_R)^3 \frac{2K_E}{(K_E \theta + 1)^3} d\theta. \quad (76)$$

In the above equations, the symbol F_f includes the consideration of particle depletion by reaction as well as elutriation, whereas, the symbol E_f is the fraction of particles escaping by elutriation with no chemical reaction occurring.

Graphical integration of Equation 76 showed that for a K of $1.56 \times 10^{-3} \text{ C}_0^{1/2} \text{ min}^{-1}$, and a θ_R of 2.0 min, 1.5 w/o of fines would escape from the bed. However, there may be some question concerning the applicability of data from a boat experiment to an individual particle. If θ_R were 10 min rather than 2 min, 7.1 w/o of fines would escape the bed for the same superficial gas velocity of 0.67 ft/sec. In any case, it does appear that with a fluorine concentration of at least 10%, the amount of escaping fines can be controlled to a low value.

5. Conclusions for Two-zone Fluorination

For the design considerations that were made, there is no indication of expected difficulties in the two-zone fluorination process.

The conclusions can be summarized as follows:

- 1) The fine uranosic oxide particles should be essentially at the same temperature as the bed during reaction, unless poor fluidization occurs, resulting in formation of agglomerates greater than 0.05 in. in diameter.
- 2) The distribution of uranosic oxide fines throughout the bed should be good, and high transport rates should prevent a build-up of fines in the lower zone.
- 3) The gas distribution of fluorine from a single injector tube is good, and an elaborate distributor would not be necessary.
- 4) For conditions in which the average fluorine concentration is 10%, over 98% of the fines would be reacted before elutriation from the bed. It is important that there be a slight fluorine excess to avoid accumulation of unreacted fines.

APPENDIX

Nomograph for Predicting the Range of Fluidizing Gas Velocities

A nomograph has been constructed to give quick estimates of the minimum fluidization velocity and the gas velocity at which the fluidized particles would be carried out of the column. This latter gas velocity is the same as the particle terminal velocity. These two velocities establish the range in which fluidization for a particular system will occur.

Rowe⁽⁴⁴⁾ has shown that the drag coefficient for a particle in a closely packed array is 68.5 times the drag coefficient for an isolated particle. Therefore, the minimum fluidization velocity can be calculated in the same manner as the terminal velocity for an isolated particle by altering the drag coefficient, C , by a factor of 68.5. A nomograph was given by Reboux⁽⁴⁰⁾ for calculating particle terminal velocities. The nomograph presented here (see Figure 72) is an alteration of Reboux's nomograph so that both the minimum fluidization velocity and the particle terminal velocity can be determined.

Illustrative Example

It is desired to know the range of gas velocities in which 0.4-mm-diameter copper shot can be fluidized by air at room temperature and atmospheric pressure. The physical properties needed for this system are

$$\rho_f = 0.0012 \text{ g/cm}^3 \text{ (gas density),}$$

$$\mu = 0.018 \text{ centipoise (gas viscosity),}$$

$$\rho_s = 8.9 \text{ g/cm}^3 \text{ (particle density),}$$

and

$$d = 0.4 \text{ mm (particle diameter).}$$

The nomograph is then used as follows:

1. The density (point 1) and the viscosity (point 2) are aligned to yield point 3 on the A scale and point 3 on the E scale.
2. Point 3 on the A scale is aligned with $(\rho_s - \rho_f)$ (point 4) on the V_{mf} side of the scale to yield point 5 on the B scale (line 3-4-5).
3. Point 5 is connected with the d scale (point 6) to yield point 7 on the CR_e^2 scale (line 5-6-7). Point 7 is the drag coefficient times the Reynolds number squared.
4. The slope of the pattern from point 7 yields point 8, which is the Reynolds number for the minimum fluidization velocity.
5. The Reynolds number is solved to give the minimum fluidization velocity by aligning point 3 on the E scale with point 8 to yield point 9 (line 3-8-9). Points 6 and 9 are aligned to yield the minimum fluidization velocity, which is 1.45 ft/sec (line 6-9-10).

Figure 72

NOMOGRAPH FOR PREDICTING THE RANGE OF FLUIDIZING GAS VELOCITIES

108-6491 Rev. 2

The terminal velocity is found in the same manner, except that the V_t side of the $(\rho_s - \rho_f)$ scale is used. Following the same procedure, the terminal velocity is found to be 23 ft/sec.

Therefore, it is estimated that the range of gas fluidizing velocities for the system is between 1.45 and 23 ft/sec.

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