RF Project..... 929

Report No..... C00-774-5

.....
FINAL

REPORT

By

THE OHIO STATE UNIVERSITY
RESEARCH FOUNDATION

1314 KINNEAR RD.
COLUMBUS, OHIO 43212

To..... U. S. ATOMIC ENERGY COMMISSION
..... Chicago Operations Office
..... 9800 S. Cass Avenue
..... Argonne, Illinois
..... Contract No. AT(11-1)-774

On..... FLUID-SOLID HEAT TRANSFER STUDY
.....
.....

For the period..... 1 May 1959 - 30 September 1964

Submitted by..... Robert S. Brodkey
.....
..... Department of Chemical Engineering

Date..... October 1964

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NOMENCLATURE

A	= area for heat-transfer
a	= surface area of solids per ft ³ of solids
c	= specific heat
C	= concentration
D _p	= average particle diameter, ft.
e	= fraction solids in transfer zone
G	= mass flow rate of gas
h	= local heat transfer coefficient between gas and particles
k _e	= effective thermal conductivity
L	= total height of bed
m	= property parameter
q	= rate of heat transfer
R	= defined by Eq. (7-2)
$\bar{u}$	= actual gas velocity in bed
T	= local gas temperature
T _i	= inlet gas temperature
T _o	= outlet gas temperature
W _g	= pound of gas
W _s	= pounds of solids
z	= vertical distance from the supporter
ε	= void fraction
α, β, γ, δ	are constants as defined in text
ρ	= density
μ	= viscosity
K	= slope of log amplitude versus z curve
φ	= phase shift

NOMENCLATURE (continued)

Φ slope of ϕ versus z curve

θ_1, θ_2 defined in Eq. (7-4)

ω frequency in radians/sec.

Nu Nusselt Number

Pe Peclet Number

Re Reynolds Number

$Z_1, Z_2, Z, P_{em}, P_{eH}, H, Y, X, m$ dimensionless parameters of the
parametric study

Subscripts

s = solid

g = gas

e = emulsion and transfer zone

b = bubble

t = time

FLUID - SOLID HEAT TRANSFER

I. INTRODUCTION

The transfer of heat between a gas and the solid particles within a packed or fluidized bed is important in estimating the temperature of a solid catalyst from the measured temperature of the gas stream. This information is essential for a kinetic analysis of chemical reactions, and for a general understanding of the transport mechanisms in such systems.

In all fluidized bed work to date using steady-state conditions, it has been impossible to measure the temperature of the solid directly, consequently there has been a number of assumptions to establish this temperature. Different opinions exist as to whether a thermocouple in a fluidized bed indicates the temperature of the fluid, solid, or some intermediate temperature. The unsteady-state method has also been used and has given heat transfer coefficients one to two orders of magnitude lower than the steady-state method. This procedure does not require knowledge of the solid temperature.

This project was directed toward obtaining heat transfer coefficients for both packed and fluidized bed systems by a frequency response method and, in addition, was directed toward obtaining direct measurements of the temperature of the solid particles within the fluidized bed.

This document is the final report on all of the work performed under Contract No. AT(11-1)-774 with the U. S. Atomic Energy Commission. It includes all work previously reported under Progress Reports 1 through 4.

Included in this introductory section are first, a general description of the fluid bed system, indicating the difficulties of rigorous analysis; and second, a discussion of particle-fluid heat transfer, which includes the general simplifying assumptions, ideal models, and rate equations.

In subsequent sections consideration will be given to theoretical models for both packed and fluidized bed systems, design of experiments to measure heat transfer coefficients by the frequency response method used in the present investigation, experimental results on packed and fluidized bed systems, and conclusions. Although this is the final report of this project under the specified contract, it is not the end of the work necessary to obtain a complete understanding of the transfer processes in packed and fluidized beds.

A. DESCRIPTION OF FLUIDIZATION

1. Types Of Fluidization

A fluidized state can be achieved by passing a fluid, at a certain velocity, through a free solid particle system bounded by a wall. The ascending velocity of fluid must be of such value that the drag force on the particles becomes equal to or greater than the gravitational force. A certain void fraction will exist. It is also possible to have the direction of fluid flow other than vertical.

There is a great difference in the mechanism and nature of fluidization, depending on whether a liquid or a gas is used. This is due to the differences in compressibility, density, and viscosity. Our interest is in gas fluidization, where the difference in density between the gas and solid is sufficiently great to assure distinct aggregate fluidization. However, it is hoped that the results of this work will be applicable to liquid fluidization (particulate fluidization).

The solids in a gas fluidized system can exist either with or without net motion. A pneumatic transport (co-current) or a continuous raining of solid particles through the ascending gas stream (counter-current) can be classified as the former. This report will be restricted to the packed bed and stationary fluidized bed which are characterized by no net solid flow and an ascending gas stream.

2. Definitions

In a fluidized bed, as the gas velocity is gradually increased, the bed will experience expansion, minimum fluidization, emulsion state, and nonhomogeneity. A bed with gas velocity less than that of minimum fluidization is characterized by stationary individual solid particles; this range is an important limiting case of the fluidized bed.

a. Minimum Fluidization and Emulsion State - The analyses and correlations of fixed bed heat transfer should be satisfactory up to the point of minimum fluidization. Reviews can be found in Leva (1) or in Zenz (2) and a more recent correlation in Gupta and Thodos (3). The visual determination in the minimum fluidized state is not difficult when approached from higher gas flows. There is no satisfactory heat transfer correlation for the emulsion state at present. This is due to the extreme sensitivity to the entrance effects and particle size distribution. Although the visual observation is not difficult, reproducibility of data presents a problem.

b. Channeling, Bubbling, and Slugging - These phenomena occur at gas rates higher than that of the emulsion velocity. Slugging tends to occur at the initiation of fluidization; however, after the particles are aerated, this effect becomes minor, especially in a larger bed. Channeling is characterized by narrow and continuous streaks of the gas stream. This can be either continuous through the entire bed length, or intermittent. Bubbles

tend to be spherical caps and appear to grow as they arise. Bubbles, channeling, and slugging hinder the effective contact of gas and solid, and cause lower conversion of heterogeneous catalytic reactions when compared to fixed bed systems. These three phenomena are extremely dependent on the entrance effects and the shape and size distribution of the particles.

3. General Approaches

Three types of approaches have been used to promote an understanding of fluidization:

- i. Fluidized bed as an expanded fixed bed and extrapolations of fixed bed data and correlations.
- ii. Fluidized bed as a well-mixed fluid.
- iii. Fluidized bed as a complex two-phase system.

The first approach was used by earlier workers and has provided some practical information concerning mass transfer characteristics of fluid beds. This approach was, and theoretically should be, successful at the flow rate below minimum fluidization. As for the second approach, various analogous properties, such as density, viscosity, temperature, or surface tension of a fluidized bed, were studied (4). Size of solid particles (in a fluidized system) relative to that of molecules, the interaction of nonuniformly shaped and sized particles, and the influence of the wall, caused the extreme irregularity and irreproducibility of data for this approach. The above two approaches provide very little basic understanding of a fluidized bed, especially at flows greater than that required for minimum fluidization.

The third approach can be divided into two categories: studies of the mechanisms of the fluid bed or parts thereof, and model studies.

Table I. Investigation on Specific Points

	Workers (Ref.)
Entrance Effect And Supporter Designs	Heertjes (5) Orgen (6) Chamberlain (7)
Bubble Rise	Baron (8) Baumgarten (9) Davidson (10) Yasui (11)
Bed Porosity	Heertjes (12)

Table I. (continued)

	Workers (Ref.)
Heat Loss	Orgen (6)
Effect of Particle Size Distribution	Fritz (13) Chamberlain (7)
Model Studies	Shen (14) May (15) Van Deemter (16)

B. FLUID-PARTICLE HEAT-TRANSFER IN A FLUIDIZED BED

At least three modes of heat-transfer may be encountered in a fluidized bed, i.e., between the bed and wall, between the bed and any submerged heat exchanger, and between the gas and solid particles. Among these modes the last one, which is considered in this report, is understood less than the other two, despite a great deal of recent work.

For the most general case, consider the cooling of a hot air stream in a fluidized bed. To aid in the discussion, all possible steps which may occur in a fluid-particle heat-transfer experimental unit are heat transfer between (1) entering gas and supporter, (2) supporter and bombarding solids, (3) supporter and gas which accompanied the solid or which resulted from gas mixing, (4) supporter and wall by conduction, and (5) gas and solids. Other steps are (6) gas-to-gas heat transfer at adjacent area by conduction or radiation, (7) transfer by gas mixing to other locations, (8) solid-to-solid heat transfer by contact or radiation, (9) transfer by solid mixing to other locations within a bed, (10) heat transfer to wall by the gas, and (11) heat loss to wall by the solids.

The above steps can be divided into four groups:

- i. The effect of the supporter (items 1-4).
- ii. The actual fluid-particle heat transfer (item 5).
- iii. Gas-gas and solid-solid transfer by conduction, radiation and mixing which must be effectively accounted for by the proper selection of the model and model parameters (such as gas mixing, solid mixing, emulsion-bubble exchange, etc.) (items 6-9).

iv. Heat loss through the wall (item 10-11).

The effect of the supporter is well demonstrated by various experiments (5, 6, 7). In an unsteady-state experiment, when the supporter has high heat capacity and high thermoconductivity, a strong discontinuity (a hump) is observed in the time-temperature curve at a point just above the supporter. Heertjes (5) studied this effect for various supporter designs and materials. The heat lost through the walls and the attempted means of evaluation of the loss will be presented later. These two effects (i and iv) are very undesirable and should be minimized.

The local heat transfer between gas and particles (item ii) cannot be studied separately because there is no way to determine the amount of heat transfer from a single particle. Further complications arise because the area may not remain constant or equal to that of other locations when the bed is not homogeneous. The value of the heat transfer coefficient for this step should be a function of the slip velocity of the gas. The correlation should be somewhat similar to that of a single particle, which is given in McAdams (17) and the modifications of Gupta and Thodos (3).

Steps 6 and 8 may be neglected in most cases because of their low driving force and high resistance. The value of q for step 5 can be evaluated only from the total heat balance and from the knowledge of steps 7 and 9. The mixing patterns of gas and solids are extremely complex, but an attempt is made to account for these in the more complex models. Minor adjustments of the mixing parameters may be used to account for the small contributions from steps 6 and 8. Actually, the parameters of mixing are used to account for effect (iii).

Previous workers have assumed simple mixing patterns of both gas and solids in order to reduce the complex problem into easily describable mathematical models. The more complex models have received attention (15, 16); they require numerical computer solutions. Independent experiments have been performed to evaluate the mixing parameters; for gas residence time studies (4), frequency response methods were used, and for solids, radioactive tracer techniques are appropriate (15).

1. Packed Bed Correlations

The objective of the packed bed heat transfer work is to gain a basic understanding at low Reynolds numbers of all the factors that affect the overall transfer of heat from the gas to the solid. The systems of particle-gas contacting for heat transfer that have been commonly encountered in the chemical engineering literature are the single-particle, the packed-bed, and fluidized systems.

Obviously single-particle heat transfer is the simplest mode and should be the most basic. A host of published works is available and these are well summarized by McAdams (17). No experimental data are available for superficial Reynolds numbers less than 10, which would be of interest for

fluidized systems. However, theoretical work has been reported by Carberry and Bretton (18). A simple theoretical analysis for the case when the Reynolds number is zero (or a single spherical particle in infinite fluid at rest) give a Nusselt number of 2.0. A further extension of this analysis reveals that the number cannot be less than two for heat transfer between air and a spherical particle in any case. Furthermore, the Nusselt number should increase as the velocity increases.

For packed-bed heat transfer, considerable published works are available. The results have been correlated by Gupta and Thodos (3). However, all available works are limited to Reynolds numbers higher than 10.

The single-particle correlation from McAdams (17), the packed-bed correlation by Gupta and Thodos (3), and the fluidized bed heat transfer results suggested by Franz (19) are included in a single Nu-Re plot for comparison (see Fig. 1).

2. General Assumptions For Fluidized Beds

(i) All previous workers assumed that the thermal properties of gas and solid are constants. Heat capacity, viscosity, density, and the thermal conductivity of gas are dependent upon variations of temperature and pressure. For a solid, only the thermal conductivity changes appreciably with temperature. In many cases, the above variables are not in the final correlation equations, except for the correlations which have dimensionless groups. The general assumption of constant properties probably would not have much effect on the results, since the variation in the results is enormous compared to these influences. Thus, the use of average values should be permissible for moderate variation of variables.

(ii) The thermal gradient within a solid particle is assumed to be very small compared to the gradient across the surface gas-film. Thus, it is assumed that the particle has a uniform temperature. When the particle has a small diameter, small heat capacity, and high thermal conductivity, this assumption is valid because unsteady-state heat transfer estimates show that the surface heat transfer is controlling, and thus, the particle temperature is constant.

(iii) In order to obtain a simple, but representative, mathematical model, some simplifying assumptions on the flow pattern or mixing of gas and solids were assumed. For the simplest cases, complete mixing or progressive flow of gas was assumed. A fixed position, definite circulating motion, or completely random mixing was assumed for the solids. Among the above three types of assumptions for solid mixing, the second seems most adequate. However, there is a tendency in the literature to use the third assumption. For the gas mixing, the actual case appears to be between the two extremes. Fritz considered both extreme cases and thought that his results confirmed the progressive assumption. Ferron (4) attempted to represent gas mixing by the use of two back-mixing parameters, which brought additional complexity to the equation.

Some workers assumed a uniform solid temperature, regardless of location in a bed; this can be justified if the mixing is extreme and random.

3. Ideal Models for Fluidized Beds

For the various assumptions of solid and gas mixing patterns, single-phase models can be constructed as in Table II.

Table II. Assumptions and Ideal Models

Solid	Gas	Used by (Ref.)
Complete mixing	Progressive	Fritz (13) Kettenring (20) Heertjes (5)
	Complete mixing	Wamsley (21)
	Back mixing	Ferron (4)
Random mixing	Progressive	May (15)

For the bed with bubbles, various workers have confirmed the suitability of the two-phase model which was first proposed by Johnston (14). The model can be pictured as follows: There are two physically distinct phases (dense and dilute) in a fluid bed when the mass flow rate is above the emulsion velocity. The dense phase is characterized by its high solid density and is continuous over the bed. The dilute phase consists predominantly of gas in discontinuous bubbles or channels. However, depending upon the assumptions which are made to describe the dense and dilute phases, the models can differ widely.

In the case of May's model (15), which seems most representative and recent, the amount of gas present in the dense phase is assumed to be equal to that of the emulsion state. The solids stay in the dense phase and mix in a random fashion, which can be described by a pseudo-diffusion constant. May also accounted for the interphase transfer by the cross-flow ratio of the gas. For gas mixing, he adopted progressive flow in both phases. In his analysis, a constant area ratio of dense to dilute at any horizontal cross section of the bed is assumed. This also implies constant average bed porosity and bubble velocity at any given cross section. The approximation of this model to the factual bed with bubbles is very rough, because many recent works on bubbles and bed porosity indicate a quite different picture (12). Thus, it may be essential to account for (1) change of bed porosity as a function of bed height and other variables, (2) bubble rise or growth during its rise, and (3) gross circulation of solids.

4. Equation for Heat Transfer

Generally, a heat transfer process can be expressed by the rate equation, $q = hA\Delta T$. In an unsteady state, all terms except A are functions of time. A detailed discussion on the definition, measurement and errors of measurement of each term in the equation will be presented next.

a. Heat-Transfer Coefficient (h) - This term is calculated from the other directly measured quantities in the equation, and its reliability is dictated by the accuracy of these measured quantities. The meaning of h depends on the process which the equation describes, and the meaning of each term. There are five distinct mechanisms in the most simple case, which has been mentioned. All of these steps contribute to the rate of particle-fluid heat transfer. If a bed is not homogeneous, additional complexity that involves transfers between phases would be introduced. Ferron combined the effects of all of the mechanisms into a single rate equation and named it the "space averaged" heat transfer coefficient.

There are three major experimental procedures. The first is simply an unsteady-state heating (or cooling) of a bed of particles. The second is more difficult and involves the design of a steady-state heat-transfer system. The third involves the use of mass transfer (drying) to provide the steady-state heat sink. In the case of the steady-state drying experiments (5, 20), if the process of drying is dictated only by the rate of heat transfer through the gas film on the particle surface, the value of the heat transfer coefficient of both steady- and unsteady-state experiments should be the same. However, there are some definite discrepancies between the two groups. For this comparison, Heertjes' values were re-evaluated and based on the total particle area. These discrepancies may be the result of inaccurate measurements of values and dissimilarity of the properties of solid particles. Ferron performed both steady- and unsteady-state experiments using identical materials and apparatus. However, his work is not sufficiently complete to provide any definite solution to this problem.

b. Area Term (A) - Two area terms have been used, namely, the surface area of the bubbles and that of the solid particles. Orgen (6) used the bubble surface area under the assumption that the major heat transfer takes place at the interphase of bubbles and emulsion. He further assumed the perfect gas law and aspherical bubble shape (based on the data by Yasui, 11). Interphase heat transfer is recognized also in the May model. The mechanism in this model is one of mass transfer, with the heat transferred as a secondary effect. The values of Orgen's heat transfer coefficient should not obey the usual dependencies of h on variables such as slip velocity, particle diameter, etc. The use of the bubble surface area is unacceptable because of the assumptions necessary for its application to particle-solid heat transfer.

The use of the solid surface area in the rate equation seems the most correct. For determination of the specific surface area of particles, various methods have been used. Some workers have assumed spherical shape, and calculated A from average particle diameter, using the simple equation

$A = 6W_s/D_{pg}$. Others used the results of screen tests, with corrections for shape factor, or used packed bed pressure drop data and Eugen's equation. Microphotography has been used to aid in determination of distribution and particle shape.

The major disagreement in the choice of the solid particle area is between the equilibrium and the total area. This difference may be the main cause of the difference in magnitudes of h shown in the results of previous workers. Thermal equilibrium between particles and surrounding gas is obtained at short distances above the supporter (within the accuracy of temperature measurements). However, there is no definite agreement on the bed height required for equilibrium.

In experiments which deal only with space- or time-averaged heat-transfer coefficients, the choice of area is not restricted because the value of q is the total heat transferred in the bed, and the terms for temperature difference are some sort of average for the height (either equilibrium or total). For the determination of the local heat-transfer coefficient, the choice of area should be that within the elementary volume under consideration.

Regardless of the choice, a constant porosity or constant particle surface area per volume is assumed in all the previous work. Heertjes (12) strongly indicates that this assumption is erroneous. The variation of porosity in the bed ranges from almost void to the value for the case of an equivalent loosely-packed bulk solid. Porosity dependence on relative location and on other variables is not well established. This complexity may be due to the solid circulation and fluctuating bubbles.

c. Amount of Heat Transferred and Heat Loss (q) - The determination of the heat transferred is very simple and direct if q represents the total amount of heat transferred, and if there are no heat losses. In contrast, for a local differential volume, the heat transferred can be attained only by indirect methods which necessitate an accurate knowledge of mixing.

The total q of drying experiments is evaluated by the amount of water evaporated (from humidities of inlet and outlet gas) and also by the sensible heat difference of the air. Account should be taken also of the desorption heat caused by unsteady inlet temperatures and humidity. However, an accurate evaluation of this term seems extremely hard in an actual case because of the uncertainty in the proportions of free moisture. Even in unsteady-state heating experiments, the desorption heat contributes significantly. If controlled inlet gas humidity is used, the effect of the heat of desorption will be small.

Some earlier experimenters neglected the effect of heat losses. Some tried to compensate for heat loss through the wall by passing hot air outside of the bed (4), by the use of heating coils, or by preheating the wall and grid. In a poorly designed unit, such as the one used by Fritz, the effect of the wall and grid is very high. Fritz's run No. 39A was recalculated by Orgen and the result indicated that the effect of the wall and

supporter corresponded to 11.3% of the heat capacity of the solid. Wamsley (21) adjusted T_i to compensate for the heat loss. Orgen (6) allowed additional solid weight to compensate for the loss. None of these means of compensation is adequate.

d. Temperature Difference (ΔT) - The temperature difference is the most troublesome term in the equation, $q = hA\Delta T$. The driving force, ΔT , must be defined so that it is consistent with the selected area, A . Except for Orgen (6), most workers took the particle surface area for A , either A_e or A_t . This means the driving force must be the temperature difference between particles and gas. Orgen assumed that the major resistance of heat transfer in the system is in the interphase between bubbles and dense phase. He assumed the solid temperature, T_s , was uniform and equal to the emulsion temperature, T_e , which was obtained from the total heat balance. In this calculation he further assumed that the bubble temperature, T_b , was equal to the exit temperature, T_o .

There is little controversy on the measurement of the inlet temperature, T_i , and the outlet temperature, T_o . However, in the precise measurement of T_o , sufficient care must be exerted to avoid the influences of bursting bubbles, non-uniform velocity, and temperature variation.

The gas temperature measurement technique, using a suction tube and a high-speed thermocouple, seems adequate. However, there are some dangers which must be avoided. When the suction is too high, it tends to disturb the flow pattern of the bed, and any solids which catch on the shield tend to change the true gas temperature. These effects are extremely hard to evaluate. Another danger is that there is no way of telling whether the gas is from the bubble or emulsion. If the suction probe is large, gases of both phases may enter simultaneously. Thus, this value of the gas temperature, T_g , must be between T_e and T_b if T_e does not equal T_b .

The most controversial term is the solid temperature. Leva (1) indicated that accurate determination of this solid temperature would solve the problems of fluid-bed heat transfer. However, without the aid of a representative ideal model this is doubtful. Contrary to Kettenring's claim, Fritz states that the reading from a bare thermocouple in the bed is that of the solid temperature. As proof he measured the temperature of the settled bed and the two readings were identical. Wamsley adopted an unsteady-state experiment to eliminate the necessity of using the solid temperature. In drying experiments, the saturation temperature of the out-going gas is used as T_s . The readings from a bare thermocouple should not indicate the true solid temperature unless a complete equilibrium between T_s and T_g around the thermocouple has been obtained. If there is no equilibrium, a bare thermocouple must indicate a value somewhere between T_s and T_g . It is practically impossible to estimate the true solid temperature from bare thermocouple readings because of the complexity of the mechanism. Fritz devised a calorimetric method to obtain the solid temperature by isolating a certain amount of solid from the bed by application of suction, and submerging the transported solids into water. This method is unacceptable because of unknown unsteady-state effects. Another method is to stop the flow

of gas intermittently and measure the temperature of the settled bed with a bare thermocouple. This value is assumed to be the solid temperature. Objections to the settled-bed measurement are the possible position change due to the bed settlement, and the time delay of measurement in an unsteady-state experiment. Wamsley reports an 8°F difference between a bare thermocouple and the settled bed, which makes the validity of using a bare thermocouple more doubtful.

The question of whether thermal equilibrium between solid and gas is obtained in a very short height of bed must be settled. Even if equilibrium is attained in experiments (5, 22), the use of A_e is questionable unless the assumption of uniform T_s is valid. If the above two assumptions are made, the excess solid ($A-A_e$) should not have any effect on the heat transfer or total q . Thus, the h based on total area should be $h_t = h_e(L_e/L) = \text{constant}/L$, where L is the bed height (corresponds to A), and L_e is the equilibrium height, corresponding to A_e . Fritz and Wamsley affirmed that $h_t = f(1/L)$, despite their widely differing basic models; however, in other experiments and in these experiments the relation is not linear. The value of A_e is very arbitrary and the assumption of uniform solid temperature is doubtful. Thus, the use of an equilibrium area, A_e , does not appear justified from the information available in the literature.

The reliability of the solid temperature measurement in most work is very questionable. Heertjes' experiments indicated that there are no radial thermal gradients of solid or gas. This is probably true in a bed with a well-designed supporter with no heat loss, with minute gross solid circulation and without bubbles. However, this is not true for most of the previous experiments. Orgen reported that the value of T_s and T_g is very much dependent upon the location in a bed, when heat losses exist. There was about an 8°F difference between the center and near the wall. Most of the previous workers assumed that the temperature of the solid is uniform because of the extreme rate of solid mixing and high rates of heat transfer. An axial thermal gradient was observed by Heertjes. Some of them have taken the log mean of the gas temperature, and some have averaged the values of T_o and T_i . This is one of the many reasons for the wide disagreement of the magnitudes of the calculated heat transfer coefficient h .

5. Additional Specific Points

The effect of entrance and static electricity on the performance of fluid bed units has been demonstrated in various experiments. Their evaluation is extremely difficult.

Entrance effects have been studied by Chamberlain, Orgen, and Heertjes. Heertjes observed a rapid depression of T_g just after the start-up in his unsteady-state experiments. He defined $e = (T_g - T_i)/(T_o - T_i)$, the fraction of sudden temperature drop to total heat loss of air when T_g is taken just above the screen; the magnitude of e is a function only of gas velocity, u_g , and the material used for the supporter. From this he concluded that heat transfer to the supporter existed. He obtained the values

of e for various materials and designs. Orgen also observed the effect of a screen on the heat transfer. He not only considered the effect of the grid as a heat transfer medium, but also the initial flow pattern which indirectly affects the heat transfer.

The effect of static electricity has been neglected in most work. When the gas is very dry, this effect is great. Contrarily, humid air lessens the problem of static electricity, but introduces some undesirable desorption heat effects.

5. Summary

The general problems, the solutions of which are essential for a better model and for an understanding of the particle-fluid heat transfer problem, include:

- (a) The definition of the heat-transfer coefficient, which involves an accurate definition and determination of the heat transferred, the area of transfer, and the temperature driving force.
- (b) The description of gas and solid mixing and gross circulation of solids.
- (c) The description of gas and solid exchange between the emulsion and bubble phases.
- (d) The description of bubble behavior and of porosity distribution.

Additional problems, which must be solved for simplicity of experimentation, interpretation of results, and improved accuracy of determined heat-transfer coefficients, are heat loss and moisture effects, and entrance effects and elimination of initial disturbances.

In this section (B) of the INTRODUCTION, we have attempted to analyze all of these problems by reviewing critically the previous work, with the hope of finding some reasons for the gross inconsistencies in the experimental results. In each subsection, certain conclusions were reached, and qualitative solutions were suggested.

Briefly, a steady-state or quasi steady-state, exclusively heat-transfer experiment should be used. To obtain accurate measurement of the heat transferred, heat losses must be eliminated. Where possible, local conditions should be considered; i.e., the local particle surface area should be used in conjunction with a local solid-gas temperature difference. The resulting heat-transfer coefficient should depend only on the particle geometry, emulsion velocity, and gas properties. Indeed, the heat-transfer coefficient becomes a criterion for the validity of the model selected. It appears that the final model will have to account for the items in the first list in this summary.

II. THEORETICAL CONSIDERATIONS

The complete physical process of fluid-to-particle heat transfer in a fluidized system involves the effects of surface transfer, gas mixing, and solids mixing. Therefore, a thorough knowledge of gas and solids mixing is essential to an accurate determination of the local fluid-particle heat transfer coefficient. Most previous workers have approached the problem by using some sort of "averaged heat transfer coefficients," which account for all three of these effects. This approach cannot provide the information necessary for the understanding of each effect separately.

In any actual heat transfer experiment, all of the measurable quantities are the result of a conglomeration of the three effects; it is impossible to separate them. Therefore, a mathematical model, which accounts for all three effects independently and at the same time describes their interdependence, is necessary. The results of this approach should provide information concerning surface exchange, solids mixing, and gas mixing; such knowledge can lead to a better understanding of fluidized systems and can be applied to such practical problems as reactor design.

In order to establish an adequate model, a discussion of each separate mechanism should be presented; a combination of these should lead to the final desired model.

A. FLUID-PARTICLE HEAT TRANSFER

Determination of the coefficient for surface transfer is the primary objective of this work. A comparison of this to the heat transfer between a single sphere and a gas can be made. In single-sphere heat transfer, the magnitude of the Nusselt number cannot be less than 2.0 according to Johnstone et al.; the Nusselt number increases as the particle Reynolds number increases. The effect of adjacent particles on the heat transfer from one particle is not well established, but it is generally agreed that the presence of neighboring particles facilitates the transfer. In a fluidized system, there are many possible effects. They are: reduction of the available area for convective transfer due to contact, removal of the boundary layer due to collision, and generation of turbulence by the motion of bubbles and collisions of the particles. All of these effects, except area reduction, facilitate transfer. Some quantitative information about the relative significance of these effects can be derived from packed-bed experiments. It appears safe to say that the local heat transfer coefficient in the bed should be greater than that of a single particle and should be subject to estimation when the deviating effects from a single particle are known.

B. SOLIDS MIXING AND GAS MIXING

A knowledge of solids and gas mixing is of extreme importance to the study of fluidized systems. In a model study, differences arise simply because of the different assumptions made in describing solids and gas mixing.

Despite their significance, very little work has been done on this subject. Romano's gas-mixing study (23) and May's solid-mixing study (15) comprise the only recent work found in the literature. Because of the limited range of study and incomplete results, one cannot obtain enough insight into the nature of these mixing problems. The presence of gas bubbles in a bed at high gas-flow rates is well established. The nature of bubbles, however, is not well established at present. The nature of solids mixing is probably more complicated than gas mixing because of the strong boundary effect of the wall, and internal circulation.

The questions which must be answered are:

1. Are gas mixing and solids mixing linear?
2. If so, what form of differential equation (linear) can represent this system?
3. If not, what kind of nonlinearity exists and what are its causes? What kind of differential (linear or nonlinear) equation can be used to approximate this system?

C. LINEAR MODELS OF PACKED AND FLUIDIZED SYSTEMS

The results of our packed-bed test runs to be subsequently described indicated that the system exhibited linear characteristics. In a fluidized system the same models might be applied to any one specific location, i.e., in effect a given point can be pictured as linear fluid bed all at the same conditions of that specific point. The assumptions necessary for a linear flow system are:

1. Constant flow properties (porosity, velocity, density, etc.).
2. Constant thermal properties.
3. Diffusional type solids mixing and conduction.
4. Diffusional type gas mixing in addition to plug flow.

Four linear flow models are possible; their simplifying assumptions (in addition to those listed above), differential equations of the system, and resulting equations for evaluation of data are summarized in Table III.

Table III. Summary of Linear Flow Models

1. Exchange Model

a. assumptions: pure plug flow of gas, no solids conduction.

b. differential equations:

$$\frac{\partial T_g}{\partial t} + m\alpha(T_g - T_s) = -\bar{u} \frac{\partial T_g}{\partial z} \quad (1-1)$$

$$\frac{\partial T_s}{\partial t} = \alpha(T_g - T_s) \quad (1-2)$$

where,

$$m_0 = \frac{c_s \rho_s}{c_g \rho_g}, \bar{u} = \text{actual gas velocity}, \alpha = \frac{ha}{c_s \rho_s}, \text{ and } m = \frac{(1-\epsilon)m_0}{\epsilon}$$

c. final equations:

$$K = \frac{1}{\bar{u}} \left(\frac{\alpha \omega^2 m}{\alpha^2 + \omega^2} \right) \quad (1-3)$$

$$\Phi = \left(\frac{1}{\alpha^2 + \omega^2} \right) \frac{\omega}{\bar{u}} (\omega^2 + (1+m) \alpha^2) \quad (1-4)$$

$$\alpha = \frac{\omega^2 \frac{m}{K\bar{u}} \pm \sqrt{\left(\frac{m\omega^2}{K\bar{u}} \right)^2 - 4\omega^2}}{2} \quad (1-5)$$

$$\alpha = \omega \sqrt{\frac{\Phi - \frac{\omega}{\bar{u}}}{\frac{\omega}{\bar{u}}(1+m) - \Phi}} \quad (1-6)$$

The values of K and Φ are the real and imaginary components of the exponent of the linear solution. These can be obtained experimentally by taking slopes of the plots of log amplitude versus vertical distance and phase shift in radians versus vertical distance.

2. Conduction Model

a. assumptions: plug gas flow with solids conduction.

b. differential equations:

$$\frac{\partial T_g}{\partial t} + m\alpha(T_g - T_s) = -\bar{u} \frac{\partial T_g}{\partial z} \quad (2-1)$$

$$\frac{\partial T_s}{\partial t} = \alpha(T_g - T_s) + \delta \frac{\partial^2 T_s}{\partial z^2} \quad (2-2)$$

$$\text{where } \delta = \frac{k_e}{c_s \rho_s (1-\epsilon)}$$

c. final equations:

$$-K^3 + 3K\Phi^2 + (K^2 - \Phi^2) \frac{m\alpha}{\bar{u}} - \frac{2K\Phi\omega}{\bar{u}} - \frac{\alpha}{\delta} K - \frac{\omega}{\delta} \Phi + \frac{\omega^2}{\bar{u}\delta} = 0 \quad (2-3)$$

$$-3K^2\Phi + \Phi^3 + (K^2 - \Phi^2) \frac{\omega}{\bar{u}} + \frac{m}{\bar{u}} \alpha 2K\Phi + \frac{K\omega}{\delta} - \Phi \frac{\alpha}{\delta} - \frac{(1+m)}{\bar{u}} \frac{\omega\alpha}{\delta} = 0 \quad (2-4)$$

3. Back-mixing Model

a. assumptions: gas back-mixing with no solids conduction.

b. differential equations:

$$\frac{\partial T_g}{\partial t} + m\alpha(T_g - T_s) = -\bar{u} \frac{\partial T_g}{\partial z} + \gamma \frac{\partial^2 T_g}{\partial z^2} \quad (3-1)$$

$$\frac{\partial T_s}{\partial t} = \alpha(T_g - T_s) \quad (3-2)$$

where γ = axial gas diffusivity.

c. final equations:

$$\gamma(K^2 - \Phi^2) + \bar{u} K = \frac{1}{\alpha^2 + \omega^2} m\alpha\omega^2 \quad (3-3)$$

$$\gamma(2K\Phi) + \bar{u} \Phi = \frac{1}{\alpha^2 + \omega^2} m\alpha^2\omega + \omega \quad (3-4)$$

4. Conduction-back-mixing Model

a. assumptions: no simplifying assumptions.

b. differential equations:

$$\frac{\partial T_g}{\partial t} + m\alpha(T_g - T_s) = -\bar{u} \frac{\partial T_g}{\partial z} + \gamma \frac{\partial^2 T_g}{\partial z^2} \quad (4-1)$$

$$\frac{\partial T_s}{\partial t} = \alpha(T_g - T_s) + \delta \frac{\partial^2 T_s}{\partial z^2} \quad (4-2)$$

c. final equations:

$$\gamma(K^4 + \Phi^4 - 6K^2\Phi^2) + \bar{u} (K^3 - 3K\Phi^2) - \left(\frac{\gamma}{\delta} + m\right) (K^2 - \Phi^2) \alpha \quad (4-3)$$

$$+ \left(\frac{\gamma}{\delta} + 1\right) 2K\Phi\omega - \frac{\bar{u}}{\delta} (K\alpha - \omega\Phi) - \frac{\omega^2}{\delta} = 0$$

$$4\gamma(K^2 - \Phi^2) K\Phi + \bar{u} (3K^2\Phi - \Phi^3) - (K^2 - \Phi^2) \omega \left(\frac{\gamma}{\delta} + 1\right) \quad (4-4)$$

$$- 2K\Phi\alpha \left(\frac{\gamma}{\delta} + m\right) - \frac{\bar{u}}{\delta} (\omega K + \alpha\Phi) + \frac{\omega\alpha(1+m)}{\delta} = 0$$

5. Mass Transfer Only Model

The mass transfer data can be analyzed by an analysis similar to that used in heat transfer.

a. assumptions: plug gas flow.

b. differential equation:

$$\frac{\partial C}{\partial t} = - \bar{u} \frac{\partial C}{\partial z} + \gamma \frac{\partial^2 C}{\partial z^2} \quad (5-1)$$

This equation is the same as Eq. (4-1) or Eq. (3-1) when $m\alpha$ (surface exchange of heat) is zero and T_g is replaced by C .

c. final equations:

$$\gamma(K^2 - \phi^2) + \bar{u}K = 0 \quad (5-2)$$

$$\gamma(2K\phi) + \bar{u}\phi = \omega \quad (5-3)$$

$$\gamma = \frac{\omega K}{\phi(K^2 + \phi^2)} \quad (5-4)$$

$$\bar{u} = \frac{\omega(\phi^2 + K^2)}{\phi(K^2 + \phi^2)} \quad (5-5)$$

or

$$\frac{\gamma}{\bar{u}} = \frac{K}{\phi^2 - K^2} = \frac{D_p}{P_e} \quad (5-6)$$

Ebach and White (24) reported a solution to Eq. (5-1) with identical assumptions. Their solution is found to be identical to the solution given above. For data analysis, however, Ebach and White, Strange (25), Liles and Geankoplis (26) and Stahl (27) used an approximate solution without the use of phase-shift data. When accurate phase-shift data are taken experimentally, the methods of analysis outlined in Part 5 of Table III are far simpler and more accurate than the method used by others. One of the original purposes of considering mass transfer was to make independent measurements of the interstitial gas velocity $\bar{u}$, and axial mixing coefficient of the gas γ , in packed and fluidized beds. When accurate values of $\bar{u}$ and γ are used in Eqs. (4-3) and (4-4), the number of unknowns is reduced by two. Thus, the analysis of the data becomes very simple.

D. SOLUTIONS OF THE LINEAR EQUATIONS

The application of a digital computer for the solution of Eqs. (4-3) and (4-4) for all the coefficients and their combinations is possible, if the values of the experimental data (K , Φ , and ω) have no error. This may be accomplished by solving a system of seven linear simultaneous equations. These data not only contain some experimental error, but also the ten coefficients must meet certain restrictions because they are combinations of α , γ , δ , $\bar{u}$, and m . It could be possible to analyze the data by taking many sets of equation systems and to calculate the most probable values of each coefficient. This would be an extravagant use of computer time; therefore, a simple way to analysis was attempted.

1. Approximate Solution

Equations (4-3) and (4-4) can be rewritten as

$$\frac{K}{\omega\Phi} + \left[\frac{1}{\omega\Phi\alpha} \right] \left[\frac{\omega^2}{\bar{u}} - 2K\Phi\omega \left(\frac{\delta}{\bar{u}} + \frac{\gamma}{\bar{u}} \right) - \delta K(K^2 - 3\Phi^2) + \frac{\gamma}{\bar{u}} \delta (K^4 + \Phi^4 - 6K^2\Phi^2) \right] \quad (4-5)$$

$$= - \left(\frac{m}{\bar{u}} \delta + \frac{\gamma}{\bar{u}} \right) \frac{(K^2 - \Phi^2)}{\omega\Phi} + \frac{1}{\alpha}$$

$$\frac{\Phi}{\omega} + \frac{1}{\alpha} \left[K + \left(\frac{\delta}{\bar{u}} + \frac{\gamma}{\bar{u}} \right) (K^2 - \Phi^2) - \frac{\delta}{\omega} (3K^2 - \Phi^2) - \frac{4\gamma\delta}{\bar{u}\omega} (K^2 - \Phi^2) 4K\Phi \right] \quad (4-6)$$

$$= - \frac{2K\Phi}{\omega} \left(\frac{m}{\bar{u}} \delta + \frac{\gamma}{\bar{u}} \right) + \frac{m+1}{\bar{u}}$$

In Eq. (4-5), the relative magnitudes of each term are such that the terms of the left side, other than the first one, can be neglected. Thus, the plot $K/\omega\Phi$ versus $(K^2 - \Phi^2)/\omega\Phi$ can provide an approximate value of α as intercept and $-\left(\frac{m}{\bar{u}}\delta + \frac{\gamma}{\bar{u}}\right)$ as the slope. Using an analogous method for Eq. (4-6),

the values of $(m+1)/\bar{u}$ and $\left(\frac{m}{\bar{u}}\delta + \frac{\gamma}{\bar{u}}\right)$ can be obtained from a plot of

$\Phi/\omega + K/\alpha$ versus $2K\Phi/\omega$ by taking the value of α obtained from the intercept. The values of $(m+1)/\bar{u}$ and $m/\bar{u}$ can be calculated from the independent measurement of superficial gas velocity $\bar{u}_0$, heat capacity ratio m_0 , and the porosity of the bed ϵ . The accuracy of the calculated value of $m/\bar{u}$ depends not only on the accuracy of m_0 , $\bar{u}_0$, and ϵ , but on the accuracy of the idealized assumption of plug flow and uniformity of the bed.

This approximate method is quite simple, and can, in addition, provide a gauge of the reliability of each measurement. If a consistent error (which is not random) exists, an analysis giving equal weight to each point is practically meaningless. An error of less than one per cent in K , ϕ , or ω can cause a much greater error in α ; even to the extent that at times a negative value of α is obtained, which is physically impossible. The approximate method should be more reliable when the particle diameter is smaller since the value of α is inversely proportional to the square of the diameter for a given Nusselt number, and the value of $\gamma/\bar{u}$ is proportional to the diameter for a given Peclet number. The shortcoming of this method is that we can not obtain the values of δ and $\gamma/\bar{u}$ separately. For larger particles the method fails because of higher values of K and ϕ , which caused the high-order terms of the equations to be no longer negligible. The two plots, ϕ/ω vs. $2K\phi/\omega$ and $(\phi^2 - K^2)/\omega\phi$ vs. $K/\omega\phi$, tend to deviate appreciably from straight lines as the particle size and gas flow rates are decreased. As an alternate approach to the data evaluation and at the same time to obtain general information with regard to the frequency responses of linear systems, a parametric study of the entire equation was undertaken.

2. Frequency Response Parametric Study

The results of parametric study can provide a means for the complete data evaluation and further information regarding the effect of each mechanism of transfer on the frequency response of the system. Also, such a study will aid in the design of frequency response experiments of similar and simpler systems, such a mass transfer or conduction through packing, where general magnitudes of parameters are known.

For this parametric study the two equations for data evaluation were rewritten in a more general form of dimensionless groups and variables.

$$Z_1 = \frac{4P_{eH}P_{em}XY^4(1-X^2) + P_{eH}Y^3(1-3X^2) + (P_{em} + mP_{eH})H^2XY^2 + HY}{(P_{em} + P_{eH})Y^2(1-X^2) + (1+m)H - XY}$$

$$Z_2 = (P_{eH} + P_{em})XY^2 + (Y/2) - [\{ (P_{em} + P_{eH})XY^2 + Y/2 \}^2 + P_{em}P_{eH}Y^4$$

$$(1+X^4-6X^2) + P_{eH}Y^3(X^2-3)X + H(P_{em} + mP_{eH})Y^2(1-X^2) - XYH]^{0.5},$$

where

$$Z = \omega D_p / \bar{u}$$

$$P_{em} = \gamma / D_p \bar{u} \quad H = 6Nu / m_o Re Pr$$

$$P_{eH} = \delta/D_p \bar{u} = k_e/c_s \rho_s (1-\epsilon) D_p \bar{u} \quad Y = \Phi D_p$$

$$m = (1-\epsilon)m_o/\epsilon = (1-\epsilon)c_s \rho_s / \epsilon c_g \rho_g \quad X = K/\Phi$$

The values of P_{eH} , P_{em} , and H represent the coefficients of the transfer: thermal conduction through the solid particles, gas back-mixing, and interfacial heat transfer, respectively. Values of P_{em} , P_{eH} , m , H , and X were assumed and a trial-and-error procedure was used to give $Z_1 = Z_2 = Z$ by varying Y . At the conclusion of the contract period covered by this final report, the study had been programed and a number of check runs completed. However, the complete range of variables had not been run so that graphical representation of the results are not as yet possible. Nevertheless, to show the usefulness of the method, the following results of an experiment on a packed-bed of 0.128-inch diameter glass beads can be used:

Experimental (29)		Parametric Solution	
Measured	$X = 0.316$	Assumed	$X = 0.316$
Measured	$Y = 0.229$	Calculated	$Y \approx 0.200$ (interpolated)
Measured	$Z = 7.25 \times 10^{-5}$	Calculated	$Z = 6.14 \times 10^{-5}$ (interpolated)
Measured	$m = 3690$	Assumed	$m = 3690$
Estimated from Literature		Assumed	
	$P_{em} = 0.532$		$P_{em} = 0.532$
	$P_{eH} = 5 \times 10^{-6}$		$P_{eH} = 5 \times 10^{-6}$
	$H = 2.48 \times 10^{-4}$		$H^* = 3 \times 10^{-4}$

*Value higher than measured might account for the slight deviation in Y and Z . Further calculations are in progress. The consistency is indicated by the close check of the measured and calculated values of Y and Z . The complete results of the parametric study will be published in theses by Kim (28) and Gates (29).

E. LOCAL APPROACH ON MODIFIED LINEAR MODEL

The differential equations of the linear model are adequate to describe the packed bed, but would have to be modified to describe the local conditions of the fluidized system by adding terms for solid circulation which are similar to the convective terms of the gas phase. The resulting equation

can be written as:

$$\frac{\partial T_g}{\partial T} + m\alpha(T_g - T_s) = -\bar{u} \frac{\partial T_g}{\partial z} + \gamma \frac{\partial^2 T_g}{\partial z^2} \quad (6-1)$$

and

$$\frac{\partial T_s}{\partial t} = \alpha(T_g - T_s) - \bar{u}_s \frac{\partial T_s}{\partial t} + \delta \frac{\partial^2 T_s}{\partial z^2} \quad (6-2)$$

where $\bar{u}_s$ is a solids circulation velocity.

By similar methods used previously (the elimination of T_s and expressions in terms of K, ϕ) the coefficients can be obtained. However, the complexity of the evaluation equation and difficulties of obtaining the necessary slopes make this approach impractical.

F. VARIOUS POSSIBLE SIMPLIFICATION OF THE TWO EQUATIONS

Many simpler models are possible; the simplest of these

$$(T_i - T_o)G c_g = W_s c_s \frac{\partial T_s}{\partial t} = ehA(T_o - T_s) \quad (7-1)$$

describe the results of frequency responses best. Other models either contradicted the results of frequency response data completely in the whole range of ω or in some partial range. The solution to Eq. (7-1) can be expressed as;

$$R = \frac{\theta_2}{\theta_1} \quad \omega = \frac{T_i - T_o}{T_o} \quad (7-2)$$

$$\frac{(T_i - T_o)}{T_o} = \text{Arctan} \frac{1}{\omega \theta_2}, \quad (7-3)$$

where

$$\theta_1 = \frac{G c_g}{ehA} \quad \text{and} \quad \theta_2 = \frac{W_s c_s}{ehA} \quad (7-4)$$

For the accurate evaluation of the angle between the vectors $(T_i - T_o)$ and T_o , a new technique was devised (28). This method can provide accuracy of the angle within a few minutes of arc.

G. MODEL STUDIES

By the use of proper approximate assumptions, various models for the fluidized bed systems can be formulated. The most reasonable model based upon the results of the temperature measurements still to be discussed and literature results previously cited as a two-zone model:

where e is the fraction of solid in the transfer zone.

The differential equation for the total heat balance is

$$G c_g (T_i - T_o) = \overleftarrow{W}_{gt} c_g \frac{\partial T_{gt}}{\partial t} + e W_s c_s \frac{\partial T_{st}}{\partial t} + \overleftarrow{W}_{ge} c_g \frac{\partial T_{ge}}{\partial t} + (1-e) c_s W_s \frac{\partial T_{se}}{\partial t} \quad (8-1)$$

and for solid phase is

$$h_t A(T_{gt}-T_{st}) + (1+e) h_e A(T_{ge}-T_{se}) = eW_{scs} \frac{\partial T_{st}}{\partial t} + (1-e)W_{scg} \frac{\partial T_{se}}{\partial t} . \quad (8-2)$$

III. DESIGN OF EXPERIMENTS

Among the various modes of heating or cooling used to cause heat transfer to or from particles to a gas, the use of a sinusoidal inlet gas temperature provides a system with a minimum of disturbances in comparison to the methods used by previous workers. Other advantages of this method are that a flexibility of operation exists, so that either transient heating or cooling experiments can be made without any additional complexity. Relative simplicity of the theoretical development and of the data evaluation are obtained, as indicated in the section on THEORY. Possible shortcomings of this method are the disturbances due to natural convection of the gas, and the interaction of the input sinusoidal disturbance with the oscillatory property of a fluidized system.

In general, all undesirable complexities and effects which are hard to describe and could introduce difficult interpretation are minimized. For instance, we have used a bed geometry of circular cross section to simplify the theoretical analysis and to obtain similarity to commercial units. A vacuum-jacketed bed container, as well as insulation around the heating zone, was used to prevent heat loss through the wall; a uniform heat flux heating unit for radial symmetry and for uniform inlet conditions was used.

As yet, no positive control of the moisture in the incoming air has been attempted because the moisture content of the available 60 psi compressed air line to our laboratory is essentially constant and very small. To minimize entrance effects, a porous stainless steel plate is used above a calming section packed with lead shot. In order to eliminate electrostatic effects the use of a gamma ray emitting isotope, a humidifier, and an anti-static agent were considered. The radioisotope activity required to eliminate the problem would be too great to be practical. The use of humid air introduces undesirable heat effects due to the possibility of absorption.

Our equipment is comprised of sinusoidal power and concentration generators, gas inlet system, testing bed, and gas-temperature and concentration measuring systems.

A. SINUSOIDAL POWER GENERATOR (see Fig. 2)

Two powerstats (Type 136) are connected in series; one is used to monitor the amplitude of input to the other powerstat, which is driven by a scotch yoke. The motion of the yoke originates from a $\frac{1}{4}$ -HP, AC motor

Figure 2 SINUSOIDAL POWER GENERATOR

(Westinghouse) connected to a Zero-Max torque converter, which in turn provides a continuous variation in the output rpm. This output is belt-connected to a speed-reducer, whose output drives the scotch yoke with two identical half-cycles (with spring actuated return). This system can generate sinusoidal power with a frequency range of 0 to 1.5 cpm and a root mean square (rms) voltage range of 0 to 100. The stability of frequency is excellent; no change in frequency was observed during eight hours of continued operation. The deviations in the rms voltage amplitude from pure sinusoidal voltage, if they exist, are very slight.

B. SINUSOIDAL HELIUM CONCENTRATION GENERATOR (see Fig. 3)

For the concentration generating unit two identical metallic bellows are connected to air and helium sources. These bellows serve as variable-volume hold-up tanks. The closed ends of the two bellows are connected to a single tie-rod which is driven by a mechanical sine motion generating device (Scotch yoke); this ensures that the bellows are 180° out of phase with respect to each other and that the total gas flow rate (sum of He and air) is always constant. The yoke drive system starts with an AC induction motor whose output is connected to a Zero-Max torque converter, which in turn is belt linked to a worm reduction gear box whose output drives the shaft of the yoke. The connection between the yoke and the gear box is made by a pair of universal joints and an extension rod because of certain space limitations. The helium is supplied from a high-pressure tank, and reduced to 50 psig before use. The air stream originates from the 60 psig compressed air line in the laboratory. The flow rates of the streams are controlled and matched by the use of two very accurate low-capacity needle valves and are checked by a wet testmeter. The flow rate of the side stream is set to 10^{-3} ft³/sec, which corresponds to an equivalent gas velocity (superficial) of 0.5 ft/min in the bed. In other words, the side stream is about 1% of the main air flow, or the helium concentration oscillates about a mean value of 0.5%. The displacement volume of the bellows is about two cubic inches. The frequency range is 0-1 cycle per second.

To generate a step input concentration of helium, which is sometimes desirable for phase measurement, a solenoid valve (normally open) in the side stream is used. The solenoid valve is actuated by one of the two poles of the micro-switch whose other pole actuates the event-marker on the recorder. Another micro-switch is used to indicate a reference point of yoke motion on the recorder.

C. TEST BED (see Fig. 4)

The test bed can be divided into three sections: calming zone, heating zone, and bed. The calming zone is made from 4.75-inch ID and 7-inch long steel tubing filled with No. 5 lead shot. Immediately above the shot is a porous stainless steel plate, which provides a uniform velocity distribution across the bed diameter. The air inlet is located at the center of closed

Figure 3 SINUSOIDAL CONCENTRATION GENERATOR

Figure 4

BED-HEATER-CALMING SECTION DETAIL

bottom. The heating zone consists of two heater plates, insulators, and gaskets. The details are shown in Fig. 2. The bed is a 4-11/16-inch ID vacuum-jacketed pyrex tube with a 400 mesh copper micro-screen supporter.

D. GAS INLET SYSTEM (see Fig. 5)

Compressed air is passed through a fiber-glass packed filter which also serves as surge tank. Two needle valves are used to give a constant pressure to the rotameter which is calibrated for air under two atm and 82°F. Air leaving the second valve goes to the calming zone. The maximum capacity of the rotameter is equivalent to a superficial gas velocity of one ft/sec in the bed.

E. GAS TEMPERATURE MEASURING SYSTEM (see Fig. 6)

The gas thermocouple unit consists of a vacuum line, probe, temperature recorder, and coordinate indicator. Vacuum is maintained in a five-gallon jar by the use of a vacuum pump. The gas flow rate through the probe is regulated by two needle valves which also provide a constant pressure at the rotameter. A schematic diagram and pertinent dimensions of the probe are given in the figure. The output signal of the thermocouple is directly fed into a recorder. A 3/16-inch stainless steel plate serves as a cover for the bed and as the probe holder, and also provides for the horizontal coordinate by sliding of the plate. The probe holder can be moved vertically within a range of 14 inches. The accuracy of positioning is better than a half a millimeter.

F. CONCENTRATION DETECTION SYSTEM (see Fig. 7)

A Gaw-Mac thermoconductivity cell is used for the measurement of the helium concentration in the gas. Some modification of the resistance bridge circuit was necessary to convert the reference gas from helium to air. A variable resistor and a needle valve for the air line are used to provide additional means of zero-point suppression for recording. The sample gas stream is taken from the vacuum line from a probe and the flow through the cell is determined by the bypass stream. A shortcoming of this detection system, when compared to that used by Romano (23), is the considerable distance between the point of sampling and the point of measurement. Possible axial mixing of fluid in the line between the two points could cause dampening of the amplitude and phase shift; the degree of this is almost impossible to analyze. Another possible error that could result in phase shift is the change of velocity of the gas as the result of pressure changes in the line. To minimize these possibilities, the shortest and smallest diameter lines possible were used. The result of a test run in a packed-bed mass-transfer experiment showed that these effects were too small to be measured. Another major difficulty was caused by the very nature of the thermoconductivity

Figure 5 GAS INLET SYSTEM

FIGURE 6. PROBE AND PROBE HOLDER

Figure 7 GAS ANALYZER-VACUUM LINE SYSTEM

cell. The output voltage of the cell is also very sensitive to the pressure in the cell and sudden changes of flow rate. For instance, when the concentration of helium was maintained constant, the output voltage of the cell would fluctuate wildly due to the pressure fluctuations in the fluidized bed. To remedy this, a surge tank (five-gallon glass jar) was connected to the line. This did not eliminate the fluctuations completely but reduced it to a tolerable level.

IV. EXPERIMENTAL RESULTS

Although frequent checking at each stage was made as construction of equipment progressed, a series of test runs was made after the completion of building to check the performance of equipment and to establish some experimental background. The test runs can be classified as empty bed, single particle, packed-bed, and fluidized bed experiments.

A. EMPTY BED HEAT TRANSFER EXPERIMENTS

The purposes of runs in the empty bed were to check the uniformity of heat flux over the cross section, the uniformity of vertical gas velocity, the effect of natural convection, and heat loss. The results indicated that the uniformity in heat flux and the gas velocity at the lower section of the bed is very satisfactory.

At a distance of 10-inches above the support, deformation of the sine wave was so great that the frequency and amplitude could not be reproduced. The height at which the wave starts to deform varies with the gas flow rate, input frequency, and amplitude of the input. At the lower gas flows, higher frequency and larger amplitudes tend to decrease the height. From these observations it can be concluded that there are strong effects of natural convection, and consequently mixing in an empty tube. This does not necessarily mean that strong effects of natural convection will exist in a packed-bed system, since viscous effects due to high surface area are much greater than buoyancy, and the actual vertical gas velocity is much higher due to a decrease in voids.

The average gas temperature in the empty bed was constant, regardless of height. This indicates that there are no net heat losses through the wall. Some dampening of the amplitude should exist in the area near the wall because of the possible transfer between wall and gas; this was detected.

B. SINGLE-PARTICLE HEAT TRANSFER EXPERIMENTS

The originally proposed solid temperature measurement technique (the

use of phosphor embedded plastic particles, optic fiber, and photomultiplier) did not materialize because the fluctuation of the local density of the bed was so gross that this fluctuation completely overshadowed the desired signal of temperature variation. Various means of filtering and integrating were tried without improvement. Thus, this method had to be abandoned. However, we still feel that the basic idea is valid and plan further use of this technique in situations where the density fluctuation is negligible. There is also the possibility that the method can be adapted to the study of local density fluctuation in fluidized bed systems. Its high frequency response and sensitivity to the density change are of particular interest. The details of this development have been published in a thesis by Sidner (30).

As an alternate solid temperature measuring device, a solder ball probe was made. This is essentially a 40 gage copper-constantan thermocouple imbedded in solder balls whose diameters ranged from 1/4 to 1 mm. To test the probe, they were used to extend the Nu-Re correlation of single spherical particles in air. The results clearly showed that heat transfer of very low flows approaches a Nusselt number of 2.0.

C. PACKED-BED HEAT TRANSFER EXPERIMENTS (PBHTE)

The packed-bed test runs were intended to test the linearity of a packed-bed system, to test the various linear flow models developed in the THEORY section, to further check the functioning of the equipment and to obtain heat transfer information about the particles under packed-bed conditions, since this is a limiting case of the fluidized bed system. Although the operational variables should be kept close to those of the fluidized bed experiments, this was impossible because of the wide difference in the gas flow rates required to produce the required conditions and because of other limitations of equipment. Therefore, an attempt was made to obtain a general understanding of the heat transfer to these particles and to evaluate the information that could be obtained from these experiments. We found additional need for work of this nature because virtually no heat transfer results exist for packed systems at the very low Reynolds number range concerned here. Some information is available on mass transfer and solid conduction in packed systems, but there is wide disagreement among the workers, thus requiring further investigation of this area.

The particles studied include glass microbeads, glass beads, plastic particles, and lead shot. No useful data on the smallest microbeads could be obtained because of the extremely low gas flow rate necessary to maintain a packed system and the sharp decay of the amplitudes which are caused by the combination of the small particle diameter and low gas flow rate. The results on plastic particles showed bad distortion of the sine wave, which was found to be caused by the difference in the time required for cooling and heating within a cycle. The causes of this distortion included variations of heat capacity with temperature, low thermal conductivity, and the

relatively large size of the plastic particles. However, the nature of this distortion is such that analysis of the data may be possible by treating heating and cooling separately, as if they were two different systems with different heat capacities.

For the purpose of illustration, the data obtained from the packed-bed heat transfer experiment G1-100 and -50 are analyzed by the approximate method of Section II-D-1. The conditions of these runs are given in Table IV. The values of K and ϕ at various values of ω are given in Table V. Plots of ϕ/ω versus $2K\phi/\omega$ and $K/\omega\phi$ versus $\phi^2 - K^2/\omega\phi$ are given in Figs. 8 and 9, respectively.

Table IV. Experimental Conditions for the
Packed-Bed Heat Transfer Run G1

Properties of G1 (glass microbeads - sample 1)

1. Material: Scotchlite glass microbeads
2. Average particle diameter: 2.12×10^{-3} ft
3. Specific heat: 0.186 or $m_0 = 1.913$

Experimental Conditions

1. Average air temperature: 100°F
 2. Measured porosity of the bed: 33.8%
 3. Superficial gas velocity:
 - 0.875 ft/sec for 100% run
 - 0.436 ft/sec for 50% run
 4. Vacuum line flow rate based upon OD of the probe:
 - 2.58 ft/sec for 100% run and
 - 1.29 ft/sec for 50% run
-

Table V. Values of K and ϕ for Packed-Bed Heat Transfer Runs of G1-100 and -50

Run	ω	100	150	200	250	300	350
100	K	4.25	8.95	15.2	21.5	28.2	35.3
	ϕ	36.5	53.0	68.5	83.0	95.7	107
50	K	21.3	36.5	49.5	62.3	75.0	
	ϕ	64.0	84.5	104	119	132.5	

In Fig. 8, the slopes of two best straight lines are 2.54×10^{-3} and 3.025×10^{-3} ; these would be the same, if the data contained no error. With estimated values of $\gamma/\bar{u}$ and δ , the effect of the neglected terms of Eqs. (4-5) and (4-6) can be shown to be too small to cause any noticeable change in the data points (less than one-tenth of one per cent). The reliability of the value of ω is quite high since the only possible source of error in this is due to the possible slippage of the chart drive; this was shown not to occur. The effects of a change in K and ϕ on the slopes of two lines are indicated in Fig. 8.

The preliminary test runs were specifically designed to study the effect of the vacuum line flow rate on the values of K and ϕ . A higher vacuum line flow rate caused an increase in the amplitude at a given point but did not change the value of K; a higher flow tended to decrease the phase shift and the value of ϕ . The value of ϕ represents a ratio of time delay between two points, one foot apart, to the time required for a cycle. A constant error in the time delay will cause the error in ϕ to increase as the frequency increases. In run G1-100, the vacuum line flow rate was set at 2.58 ft/sec, which was based on the measured porosity of 33.8 per cent. However, the porosity calculated from the experimental value of $(m+1)/\bar{u}$ gave 38 per cent, i.e., from $\frac{m+1}{\bar{u}} \approx \frac{m}{\bar{u}} = \frac{m_0}{u_0} (1-\epsilon)$. Therefore, the vacuum line flow

rate should have been 2.3 ft/sec rather than the 2.58 ft/sec used. Based upon this fact and the result of the test runs, the values of ϕ should be greater than that given in Table V and the error should be greatest for an ω of 350 rad/hr. To illustrate the correction, the lowest two points (350 rad/hr) in Fig. 8 can be considered. The amount of correction necessary to obtain the same slope for both lines amounts to only a one per cent increase in ϕ . The slopes in Fig. 8 agree well. All the necessary results $\left(\alpha, m/\bar{u}, \epsilon, \frac{m}{\bar{u}} \delta + \frac{\gamma}{\bar{u}} \right)$ are given in the figure.

The result of this experiment can be compared to the available data in literature. The values of the heat transfer coefficient (Nusselt number) agree well with the results given by Gupta and Thodos (3) (see Fig. 1, solid

Figure 8 RESULTS of PBHTE GI 100

Figure 9 RESULTS of PBHTE G150

circular points). Furthermore, these results are consistent with our premise that "the Nusselt number cannot be less than 2.0 for air to spherical particles." Unfortunately we cannot obtain separate values of $\gamma/\bar{u}$ and δ from

our result as previously mentioned. However, the values of $\left(\frac{m}{u} \delta + \frac{\gamma}{u}\right)$

provide some information. For axial back-mixing of a liquid in packed beds, Liles and Geankoplis (26) reported a Peclet number of 0.32 and Ebach and White (24) reported 0.4. These correspond to values for $\gamma/\bar{u}$ of 6.625×10^{-3} and 5.3×10^{-3} , respectively, and they are greater than the experimental value for $\left(\frac{m}{u} \delta + \frac{\gamma}{u}\right)$ of 2.8×10^{-3} , even if δ is negligible. It seems

quite obvious that the axial mixing of gas flow in a packed bed must be different from that of liquids as has already been established by others from mass transfer experiments. The only work available for gas back-mixing is that of McHenry and Wilhelm (31) which indicates a Peclet number of 1.88 and which gives $\gamma/\bar{u}$ of 1.13×10^{-3} ft for the G1 particles; this is consistent with the value of $\left(\frac{m}{u} \delta + \frac{\gamma}{u}\right)$ (2.8×10^{-3}), since it is less.

If we use the value of $\gamma/\bar{u}$ given by McHenry and Wilhelm and the experimental values of $m/\bar{u}$, the values of δ can be calculated. They are 4.48×10^{-3} for run 100 and 5.28×10^{-3} for run 50. No measurements of δ , which represents the thermal diffusivity of a packed bed in a vacuum, have been made. However, some work is available on the value of effective thermal conductivity of a packed bed when the Reynolds number approaches zero. Willhite et al. (32) reported the radial effective thermal conductivities of packed beds using various gases and a liquid. Fortunately the particles they used are similar to ours (particle diameter of 1.85×10^{-3} ft compared to our 2.12×10^{-3} ft). Since the thermal conductivity of a packed bed of small spherical particles should be independent of direction, the values given by Willhite et al. for the radial direction should be the same as that for the axial direction. To calculate the value of the thermal conductivity the bed would have in a vacuum, a linear contribution of the gas and solid was assumed. In other words, the values given by Willhite et al. were expressed as the sum of the contribution of the solid, which should be constant, and the contribution of the gas, which is the product of a constant and the thermal conductivity of the gas. The most probable value of the contribution of the solid for the four different fluids is 0.0805 Btu/(hr)(ft)(°F). The maximum error is 10 per cent. This value can be converted to δ by using the following relation:

$$\delta = \frac{k_e}{c_s \rho_s (1-\epsilon)}$$

where ϵ is 37 per cent and $c_s \rho_s = 32.8$. Thus, δ is 4.015×10^{-3} ft²/hr which can be compared to 4.48×10^{-3} for run G1-100, and 5.28×10^{-3} for run G1-50. In a packed bed the value of δ should increase as the void fraction decreases because fewer voids will create closer contact of particles. This is the

case for our experimental results. Since the particle size distribution will probably cause a difference in the pattern of contact among the particles and since these effects on the values of δ are not yet established, theoretically or experimentally, a detailed discussion is not possible. However, these three values are in good agreement.

Unfortunately the approximate method of solution can only be used for analysis of the smaller glass microbeads. For all the other data taken, the total parametric study is necessary. At the termination of the contract period the parametric study had just become available and no time was available for the reduction of the other packed-bed data. The actual experimental results for the additional runs and evaluation in terms of the parametric study can be found in theses by Kim (28) and Gates (29), to be published shortly.

D. PACKED-BED MASS TRANSFER EXPERIMENTS (PBMTE)

In packed-bed mass transfer experiments our experience and some theoretical analyses based on published correlations revealed that it is impossible to obtain results in the range of experimental variables in which we are interested. The superficial Reynolds number is less than ten. In this range, the Pecklet number $Pe = D_p \bar{u} / \gamma$, is nearly constant and varies from 0.32 to 2.0 depending upon the system (24, 25, 26, 27, 31). This means the values of $\gamma / \bar{u}$ should be of the order of one-thousandth in the unit of ft^{-1} . In other words, to obtain a measurable amount of amplitude dampening at one cps, the bed height should be at least 100 feet. The fact that in all packed-bed mass transfer runs, no dampening of amplitudes in the sinusoidal inlet experiments and no deformation of the step input in those experiments was observed at the various heights in the bed, indicates that axial mixing in the flow line is negligible, or at most is of a magnitude comparable to the packed-bed system of small particles. The same results of course are not expected in the fluidized bed mass transfer experiments.

E. FLUIDIZED BED MASS AND HEAT TRANSFER EXPERIMENTS (FBMTE and FBHTE)

A series of fluidized bed heat transfer experiments were conducted to study the general qualitative nature of the system. These were used to test the linear flow models and to develop a theoretical model for the description of the system. In addition, two sets of mass transfer experiments were undertaken to determine the general degree of gas back-mixing in the system.

When the fluidizing gas velocity is low so that only infrequent and discrete bubbles are observed in the bed, the usual plots of log amplitude decay and phase shift against the distance parameter, z , show a strong similarity to that obtained in the packed bed experiments; i.e., log A versus

z and ϕ versus z gave straight lines with slopes of similar order of magnitude. There is a small scatter of individual waves about some mean values as the bubbles pass by the tip of the probe. This becomes greater as the gas flow rate and the bubble frequency increases. Finally, when gas flow rate is increased to the extent that a violent motion of solids and bubbles is observed, the lines of the two plots are quite different (see Figs. 10 and 11). These results indicate that the dampening of amplitudes and the phase shift occur within 0.2-inch from the screen and that a near or at equilibrium solid and gas temperature exists in the rest of the bed. This must be caused by the violent circulation and mixing of solid particles in the equilibrium section of the bed. There is little hope that a simple linear model will be adequate or even one with variable ϵ and a solid circulation term.

The foregoing results of gas temperature measurements in the fluidized bed showed that it could be divided into two characteristic zones; i.e., a decaying zone (the first 1/4-inch of the bed) and a constant gas temperature zone (the rest of the bed). In the lower portion of the decaying zone, the waves are very sinusoidal but tend to be more distorted as the distance from the support is increased. The distortion and the scatter is most pronounced at the area near the interface of the two zones. In the decaying zone, the recorded amplitude of the gas temperature was strongly affected by the probe suction rate, the higher suction rates giving higher amplitudes.

In the constant gas temperature zone, the recorded sine waves had a slight superimposed random scatter but they were very stable and reproducible. Changes in suction rate did not affect the wave in this zone. Identical waves were recorded everywhere in this zone and were equal to the exit gas temperature wave. This was taken as an indication of uniform gas and solid temperatures in this zone. If the mixing rate of gas is extremely high, a minute but uniform driving force (temperature difference between gas and the solid) could exist. In order to substantiate the results of the suction probe, use was made of mass transfer experiments, of the bare thermocouples, of the solder thermocouple probe, and of a copper screen thermocouple. The results of the mass transfer experiments indicated that waves were very sinusoidal near the bottom of the bed but became very irregular and irreproducible as the distance from the supporter increases. The height at which a noticeable deformation started corresponded to that in the gas temperature. A bare 40-gage thermocouple inserted into the bed gave similar results to those of the suction thermocouples, i.e., a rapid decay of amplitude near the screen and the equal temperature in the rest of the bed. Two solder ball thermocouples which have widely varied time constants were inserted in the upper section of the bed. Both thermocouples indicated identical temperature, thus indicating an extremely high rate of interfacial transfer. These thermocouples did show a slightly different temperature in the decaying zone. The recorded temperature of the supporting copper screen was little different from that of the reference thermocouples. This and the results of solder ball experiments tend to confirm the findings of Bakker and Heertjes (33), i.e., the presence of high porosity near the screen. The particles near the screen may be less mobile and less dense because of the jetting air from the openings of the screen and the limitation imposed by the screen. From these

Figure 10 $\ln A$ vs. z for FBHTE G2 60

Figure 11 ϕ vs. z for FBHTE G2 60

various observations, we feel that the fluidized bed can be divided into two zones: transfer and near equilibrium (a small but finite Δt may exist in this zone).

At the termination of this contract period, further evaluation of the fluidized bed runs in terms of the theories presented in Sections II-F and G has not been made. They will appear in the thesis of Kim (28).

V. CONCLUSIONS

The feasibility of the use of a frequency response method for the determination of the particle-to-gas heat transfer in packed and fluidized bed systems has been demonstrated. For packed beds there is also the possibility of determining the axial diffusion or gas back-mixing and the effective thermal conductivity of the bed from the same experiments. To this end, an approximate solution for small particles and a general parametric solution of the controlling equations was formulated and preliminary results from the study cited. Comparisons of the theoretical results with experimental runs showed good agreement, and in effect was a determination of the parameters controlling the heat transfer in the system (particle-to-gas transfer, back-mixing, and conduction). The general parametric study will be useful in guiding other frequency response experiments involving some or all of these mechanisms. Although not completed in time for this final report, it is expected that confirmation and extension of the presently available packed-bed correlations will be possible when the final evaluation of all of the data is completed.

A series of fluidized bed experiments was completed. The experiments have in part been evaluated so as to obtain some ideas on the manner in which heat is transferred in such systems. Based on these results, a number of theoretical models have been described. The most promising models are a single-zone, completely mixed system, and a two-zone model involving a lower, small transfer zone and an upper, large, near-equilibrium zone. However, a complete evaluation with the available data has not been completed at this time.

As mentioned in the INTRODUCTION the work under this contract has resulted in useful information about the mechanism of the transfer of heat in both packed and fluidized beds, but until all the available data (and further data that may be necessary in the future in light of these results) have been processed, one cannot with complete confidence say that an understanding of the transfer processes in packed and fluidized beds has been obtained.

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