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Modeling of Fluidized Bed Silicon Deposition Process

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June 15, 1977

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PREFACE

The work described herein was performed by the Control and Energy Conversion Division of the Jet Propulsion Laboratory.

DEFINITION OF SYMBOLS

C_A	concentration of gas A, g·mol/cm^3
C_d	drag coefficient
C_{Ab}, C_{Ac}, C_{Ae}, C_{Ag}	concentration of A in the gas bubble, in the cloud-wake region, in the emulsion phase, and at the particle surface, respectively, g mol/cm^3
$\bar{C}_{Ab}$	average concentration of A in the bubble, g mol/cm^3
$\bar{C}_A$	average concentration of A in the bed
C_{Ai}	concentration of A in the entering gas stream, g mol/cm^3
D	molecular diffusion coefficient of gas, cm^2/sec
d_b	effective bubble diameter, cm
d_p	particle diameter, cm
dp	surface mean particle size, cm
d_t	tube diameter, cm
F_0, F_1, F_2	feed rate of solids, outflow rate of solids, and carryover rate of solids by entrainment, respectively, gm/sec
Fr_{mf}	Froude number at minimum fluidization condition, U_{mf}^2/d_pg, dimensionless
g	980 cm/sec^2, acceleration of gravity
h	height, cm
$(K_{be})_b$, $(K_{be})_e$, $(K_{be})_f$	overall coefficient of gas interchange between bubble and emulsion based on volume of bubbles, volume of emulsion, and total volume of bed, respectively, sec^{-1}
k	entrainment coefficient
k_d	mass transfer coefficient between fluid at a particle, cm/sec
k_d	mass transfer coefficient in fluidized bed, sec^{-1}
$(k_d)_b$	overall mass transfer coefficient for bubbling bed, sec^{-1}
$(k_d)_{mf}$	mass transfer coefficient between fluid and a particle at minimum fluidization, cm/sec
k_{dt}	mass transfer coefficient for particles in bubbles, cm sec^{-1}

ℓ	height, cm
L_f	height of a bubbling fluidized bed, cm
L_{mf}	bed height at minimum fluidization condition, cm
M_w	molecular weight
η	bubble frequency, sec^{-1}
N_A	number of moles of A
N_{Ab}	number of moles of A in bubble
ΔP	pressure drop, $gm \cdot wb/sm^2$
P_0, P_1, P_2, P_b	size distribution of feed solids, outflow solids, entrained solids, and solids in the bed, respectively, cm^{-1}
r_0	particle radius, cm
r_p, min_f	minimum particle radius in feed particles, cm
r_p, max_f	maximum particle radius in feed particles, cm
R	half the distance between particles in particulate bed or emulsion, cm
Re_p	particle Reynolds number, $d_p u_0 \rho_g/\mu$, dimensionless
Re_{mf}	particle Reynolds number at minimum fluidization condition, $d_p u_{mf} \rho g/\mu$, dimensionless
Sc	Schmidt-number, $\mu/\rho_g D$, dimensionless
Sh	Sherwood number, $k_d d_p/D$, dimensionless
Sh_t	Sherwood number for particles in bubbles, $k_{d,t} d_p/D$, dimensionless
Sh_{mf}	Sherwood number for particles in emulsion or at minimum fluidization conditions, $K_{d,mf} d_p/D$, dimensionless
U_b	velocity of a bubble rising through a bed, cm/sec
U_{br}	velocity of a bubble with respect to the emulsion phase, cm/sec
U_f	upward velocity of gas at minimum fluidizing conditions, cm/sec
U_{mf}	superficial fluid velocity at minimum fluidizing conditions, cm/sec
U_t	terminal velocity of a falling particle, cm/sec

U_0	superficial fluid velocity (measured on an empty tube basis) through a bed of solids, cm/sec
V_b	volume of a gas bubble, cm^3
W	weight of solids, g
α	specific surface or surface of solid per volume of bed, cm^{-1}
α'	specific surface or surface of solid per volume of solid, cm^{-1}
γ_b	ratio of solids dispersed in bubbles to the volume of bubbles in the bed, dimensionless
γ_c	ratio of solids in the cloud-wake region to volume of bubbles in the bed, dimensionless
γ_e	ratio of solids in the emulsion to volume of bubbles in the bed, dimensionless
δ	fraction of fluidized bed consisting of bubbles, dimensionless
ϵ	void fraction, dimensionless
ϵ_e, ϵ_f, ϵ_{mf}, ϵ_m	void fraction in the emulsion phase of a bubbling bed, in a bubbling bed as a whole, in a bed at minimum fluidizing conditions, and in a packed bed, respectively, dimensionless
μ	viscosity of gas, g/cm•sec.
ρ_g, ρ_s	density of gas and solid, respectively, g/cm^3
ϕ_s	sphericity of a particle, dimensionless

ABSTRACT

Modeling of the fluidized bed for silicon deposition is described. The model is intended for use as a means of improving fluidized bed reactor design and for the formulation of the research program in support of the contracts of the Silicon Material Task for the development of the fluidized bed silicon deposition process. A computer program derived from the simple modeling is also described. Results of some sample calculations using the computer program are shown.

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SECTION I

INTRODUCTION

The Low-Cost Silicon Solar Array (LSSA) project sponsored at JPL by ERDA has the responsibility for developing the technology and the industry-capability for producing low cost solar arrays. The objective is the achievement of a production capacity of 500 megawatts (peak) at a cost of $500 per kW by 1986. The program of LSSA is divided into four technology tasks, each of which has the responsibility for advancing a particular technical area so as to achieve the production and cost goals. The first of these tasks is the Silicon Material Task, for which the 1986 objective is to reduce the price of Si material, suitable for the fabrication of solar cells having satisfactory performance, from the present price of about $65/kg (for semiconductor grade Si) to less than $10/kg.

To achieve the price objective of the Silicon Material Task, new areas of chemical processing and solar cell technology must be studied and developed, since no modification of the basic processes presently used commercially will lead to the attainment of the $10/kg price. The inability of the present commercial process to meet the task objective is revealed from a consideration of the features of this process. The first step is the reaction of metallurgical grade Si, having a purity of about 98.5%, with HCl in a fluidized bed reactor to yield the intermediate $SiHCl_3$ in a mixture of chlorinated silanes. After chemical treatment and fractional distillation to obtain very pure $SiHCl_3$, semiconductor grade Si is produced by chemical vapor deposition resulting from the H_2 reduction of $SiHCl_3$. This method of producing extremely pure Si is costly and energy intensive, the final deposition consuming an estimated 385 kWh/kg Si product.

Several different chemical processes are being investigated under JPL contracts in the task program. Two of these, at Union Carbide and Battelle, incorporate fluidized bed reactor technology. The contract with Union Carbide involves the hydrogenation of $SiCl_4$ to $SiHCl_3$ in a fluidized bed as well as a process for the deposition of Si from SiH_4 in another fluidized bed. In the contract at Battelle the process is for the Zn reduction of $SiCl_4$ in a fluidized bed reactor. These two efforts are presently the most advanced in the task program and are being appropriately emphasized.

To support the developments in fluidized bed technology, which is the basic element of these two important contracts, the task program includes an in-house JPL subtask for studies of fluidized bed reactor technology as well as a subtask for consultation in this area by Professors O. Levenspiel and T. Fitzgerald of Oregon State University. The in-house program consists of efforts in modeling of the deposition of Si in a fluidized bed reactor, experimental studies of SiH_4 pyrolysis and fine particle fluidization, and thermodynamic and chemical engineering analysis of the reactions in the fluidized process for silicon production. The initial phase of the effort in modeling is described in this report.

SECTION II

APPROACH

The purpose of this study is to develop an analytical model of a particular chemical reaction--the pyrolysis of SiH_4--occurring in a fluidized bed reactor. The model is intended for use in determining guidelines for reactor design and for the structuring of experimental investigations to achieve system optimization. The methodology of this study is (1) to specify the system, (2) to describe the particular and general assumptions, (3) to state the appropriate physical laws to be used at each step, (4) to apply the limitations of the system characteristics and assumptions in the derivations of the mathematical expressions, and (5) to develop a suitable computer program. To facilitate the systematic development of the computer program, this report is divided into sections for (1) a general description of the process, (2) modeling of the gross bed behavior, (3) modeling of the particulate bed, (4) modeling of the bubbling bed, (5) a description of gas interchange between bubble, cloud, and emulsion, (6) an analysis of mass transfer rate to solid particles in a bubbling bed, (7) particle growth rate in a particular bed, (8) particle growth rate in a bubbling bed, (9) entrainment, (10) overall mass balance of the bed, and (11) results, conclusions, and recommendations.

This paper deals with the fluidized bed silicon deposition process designed to eliminate the difficulties associated with the current state-of-the-art. In Figure 2-1, a schematic diagram of a fluidized bed is shown. From the bottom left-hand side, small feed-particles of silicon are introduced while the feed gas of silane and hydrogen (carrier gas) comes in from the bottom of the bed. The silane is pyrolyzed at the surface of the silicon particles, the resulting Si deposits causing the increase in size of the seed particles, which are eventually removed as the product. Unreacted silane and hydrogen gas escape through the top of the bed concurrently elutriating small amounts of small-size particles. This process is continuous and has the advantage of using a very large reaction surface area to produce pure silicon on a much larger scale than the conventional chemical vapor deposition (Siemens) process.

Our problem can be stated as follows: Given a set of operating conditions, which include the bed size, the feed particle characteristics, and the feed gas characteristics, determine (1) the type of fluidization (i.e., whether it is bubbling or particulate type), (2) the optimum gas flow rate, (3) the power requirement, (4) the silicon particle growth rate, (5) the entrainment rate, and (6) the deposition rate of Si.

Several assumptions and a number of limiting conditions were employed in the modeling of this process. They are as follows:

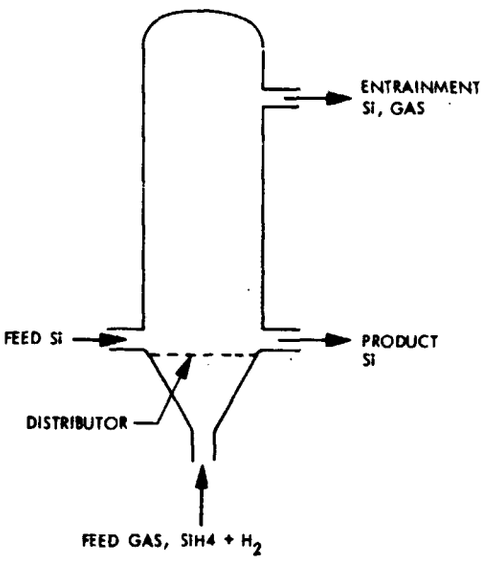

Figure 2-1. Fluidized Bed Concept for Silicon Production

(1) At temperatures above 1000°C, the primary reaction is the heterogenous pyrolysis of SiH_4 on the surface of the silicon particles.

The rates of adsorption and reaction are much faster than the transport of the reactant species, SiH_4, to the reaction zone (the hot surface), and thus the growth rate of the particle size is diffusion-limited.

(2) The bed is either bubbling or particulately fluidized. In other words, the fluidization of particles smaller than 50 μ, which would cause abnormal fluidization, will not be considered.

These two types of fluidization are schematically shown in Figure 2-2. For larger particles, bubbles moving up are observed (Figure 2-2a), and for smaller particles (but still larger than 50 μ) uniform fluidization is observed (Figure 2-2b). This uniform phase in the particulate bed and the bubbling bed (outside the bubbles) is called the emulsion.

The basic flow chart for the modeling is given in Figure 2-3. As shown here, first, the set of input parameters is given. From this, the gross behavior of the bed is obtained including the operating bed weight, the pressure drop across the bed, the distributor requirement and the type of fluidization. If the bed is particulate fluidized, the mass transfer rate of gas onto solid particles and thus the particle growth rate can then be obtained relatively simply. If the bed is bubbling, the bubble and the emulsion characteristics must be studied separately with the aid of pertinent experimental data. Then the interaction between the two phases must be investigated to determine the mass transfer rate of the gas onto the solid particles in order to permit the calculation of the particle growth rate as a function of the particle diameter.

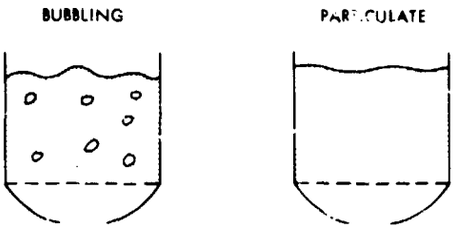

Figure 2-2. Types of Fluidization

When this growth rate is obtained, the mass balance of silicon particles in the feed, product, entrainment streams and the bed is then calculated. The particle size distribution of the bed and the deposition rate can also be determined.

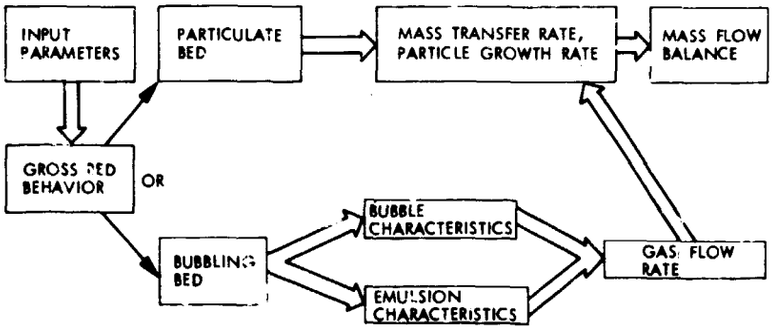

Figure 2-3. Flowchart for Modeling

SECTION III

MODELING OF GROSS BED BEHAVIOR

The input parameters necessary for the definition of the fluidized bed are as follows: feed particle size distribution $P_0(d_p)$; production rate F_1; sphericity of particles ϕ_s, which is assumed to be uniform regardless of the particle size; void fraction in packed bed ϵ_m, as a function of ϕ_s (Ref. 3-1); voidage at minimum fluidizing condition ϵ_{mf}, and as a function of ϕ_s; gas density ρ_g; solid density ρ_s; viscosity μ; diffusion coefficient D; concentration of silane in the feed gas C_{Ai}; bed weight W; and the bed diameter d_t.

In order to estimate the bed characteristics, it is necessary to know the average particle size in bed. However, without the knowledge of the bed particle size distribution $P_b(d_p)$ this is not possible. Thus, at the beginning, $\bar{d}_p$ is obtained from P_0. This will be replaced by $\bar{d}_p(P_b)$ later. Since the important parameter in the bed is the ratio of surface area to volume of solid particles, which is inversely proportional to d_p, the average particle size is obtained as the inverse of the average of the inverse of the particle size. That is,

$$\bar{d}_p = \frac{1}{\int \frac{P_b(d_p)}{d_p} \, d(d_p)} \tag{1}$$

Using the above information, the gross behavior of the fluidized bed is then predicted as follows:

(1) Bed height at minimum fluidizing conditions: There is a certain velocity of gas for the given bed below which the solid particles are packed together and no fluidization is observed. Above this velocity, the particles are moving around in the bed with gas to create a fluid-like phase in the bed. This velocity is called the minimum fluidizing velocity, and the requirement to reach this velocity is called the minimum fluidizing condition.

$$L_{mf} = \frac{W}{d_t^2} \frac{4}{\pi} \frac{1}{(1 - \epsilon_{mf})(\rho_s - \rho_g) g} \tag{2}$$

where g is the gravitational constant.

(2) The pressure drop across the bed is obtained by balancing the pressure loss with the bed weight.

$$\Delta_p = \frac{W}{d_t^2} \frac{4}{\pi} \tag{3}$$

(3) The superficial fluid velocity at minimum fluidizing conditions U_{mf} can be obtained by balancing the pressure loss with either the viscous force for the small-particle-size bed or the kinetic force for the large-particle-size bed. For an assumed value for the superficial fluid velocity of the bed U_0, then

$$U_{mf} = \frac{\bar{d}_p^2 (\rho_s - \rho_g) g}{1650 \mu} \text{ when } Re_p \equiv \frac{\bar{d}_p \rho_g U_0}{\mu} < 20 \tag{4}$$

or

$$U_{mf}^2 = \frac{\bar{d}_p (\rho_s - \rho_g) g}{24.5 \rho_g} \text{ when } Re_p > 1000 \tag{5}$$

If $20 < Re < 1000$, an interpolated value of U_{mf} would be used.

The superficial fluid velocity of the bed U_0 can be estimated from this information. If a vigorously mixing bed is assumed for maximum silicon production, then, according to literature observations (Reference 3-1):

$$U_0 > 2U_{mf} \tag{6}$$

However, there is an upper limit for U_0, since the U_0 is larger than the terminal velocity of the particles, then

$$U_t = \left(\frac{4 g d_p (\rho_s - \rho_g)}{3 \rho_g C_d} \right)^{1/2} \tag{7}$$

where d_p is the minimum particle size in bed and C_d can be obtained empirically; then some particles are lost through entrainment. This tendency of entrainment would be increased as U_0 increases.

(4) The type of fluidization of the bed can be determined by an empirical formula (References 3-2, 3-3) under normal fluidization conditions. If the product of four dimensionless groups

$$Fr_{mf},\ Re_{\rho mf},\ \frac{\rho_s - \rho_g}{\rho_g},\ \frac{L_{mf}}{d_t}$$

is larger than 100, the bed is bubbling; otherwise, the bed is particulately fluidized. Here

$$Fr_{mf} = \frac{U_{mf}^2}{\bar{d}_p g}, \tag{8}$$

$$Re_{\rho mf} = \frac{U_{mf} \cdot \bar{d}_p \cdot \rho_g}{\mu} \tag{9}$$

SECTION IV

PARTICULATE BED

If the bed is particulately fluidized, which is depicted in Figure 4-1, the mass transfer rate of gas onto the solid particles is obtained in the following way (Ref. 4-1). The individual particles of radius r_0 are considered to be separated from other particles by a distance of 2R, where R can be derived easily from the voidage of the bed:

$$R = \frac{r_0}{(1 - \epsilon)^{1/3}} \tag{10}$$

Gas enters from the bottom with the velocity U_0. The amount of gas transferred to the individual particle surfaces by diffusion, as well as by this forced convection, is to be calculated.

This is a very complex two-dimensional flow problem for each particle. The description can be approximated by use of the penetration theory as follows. Assume that at t = 0, the concentration of the silane at r = R is the same as that of the incoming gas, that at $r = r_0$, the concentration is zero, and that the diffusion which is one-dimensional in the radial direction only proceeds until t = T. At t = T, the original situation is reestablished and the ensuing diffusion process is repeated again. This is repeated at every mT interval where m is an integer. The period T is proportional to the ratio between the incoming velocity U_0 divided by particle radius r_0.

The result is summarized in a formula for the mass transfer coefficient, k_d,

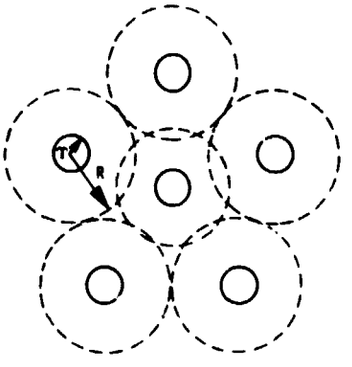

Figure 4-1. Particulate Bed

$$Sh = \frac{k_d d_p}{D} = \frac{2 + \left[\dfrac{2\zeta^2 (1 - \epsilon_f)^{1/3}}{[1 - (1 - \epsilon_f)^{1/3}]^2} - 2\right] \tanh \zeta}{\dfrac{\zeta}{1 - (1 - \epsilon_f)^{1/3}} - \tanh \zeta} \tag{11}$$

where

$$\zeta = \left[\frac{1}{(1 - \epsilon_f)^{1/3}} - 1\right] \times 0.3 \, Re_p^{1/2} \, Sc^{1/2} \tag{12}$$

$$Re_p = \frac{d_p U_0 \rho_g}{\mu}$$

and

$$Sc = \frac{\mu}{\rho D}$$

At the limit when $R \rightarrow \infty$,

$$Sh \rightarrow 2 + 0.6 \, Re_p^{1/2} \, Sc^{1/3} \tag{13}$$

which is known as a single-sphere diffusion formula. This result will be used later in the bubble phase analysis.

SECTION V

LUBBLING BED

To obtain the mass transfer rate of gas in the bubbling phase it is necessa y to understand the bubble and the emulsion characteristics first. From the literature (Reference 3-1) it is noted that the emulsion phase behaves very much like a bed at minimum fluidizing condition. Thus, the upward velocity of gas in the emulsion phase is defined as U_f. Then, by assuming that the emulsion phase behaves approximately according to the minimum fluidizing condition, one obtains,

$$U_f = \frac{U_{mf}}{\epsilon_{mf}} \tag{14}$$

The bubble phase is analyzed as follows. First the bubble diameter d_b is defined assuming that d_b is constant throughout the bed (although this is not true in the beginning of the bed), and that the bubble frequency passing by a certain point in the bed is n. Then the velocity of the rise of bubbles U_{br} is similar to the velocity observed in the system of a bubbling liquid of low viscosity, in which case inertial forces predominate. By equating the inertial force to the gravity force, the relation for a single bubble is obtained:

$$U_{br} = 0.711\ (gd_b)^{1/2} \tag{15}$$

For bubbles rising together in the bed, the absolute velocity would be, then,

$$U_b = (U_0 - U_{mf}) + U_{br}. \tag{16}$$

Depending on the bubble velocity there are two types of bubble formations as shown in (Figure 5-1).

If $U_b > U_f$, a cloud will form around the bubble (Figure 5-1a) in which the gas circulates and never escapes to the outside. The thickness of the cloud decreases as U_b becomes larger. For vigorously bubbling beds, where $U_b > 5U_f$, the cloud thickness becomes negligible. If $U_b < U_f$ (Figure 5-1b), no cloud forms and the gas passes through the bubble as a shortcut to the top of the bed. The higher mass transfer rate is obtainable for the first case; i.e., $U_b > U_f$ and this condition will be used.

The superficial velocity U_0 can be expressed as follows:

$$U_0 = (1 - \delta)\ U_{mf} + \delta U_b \tag{17}$$

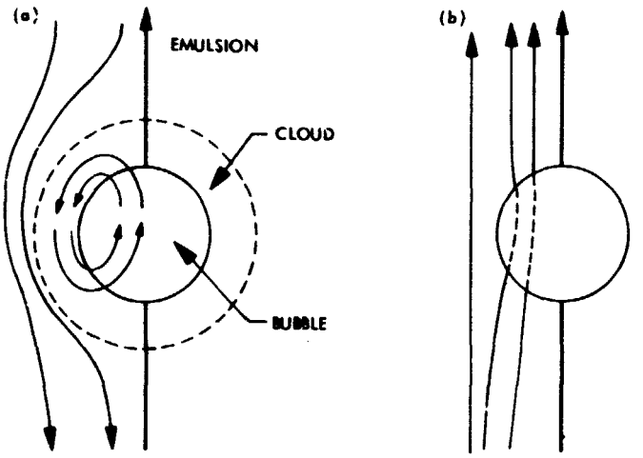

Figure 5-1. Bubbles

where δ is the volume fraction of bubbles in the bed. The first term is the velocity contribution from the emulsion phase and the second is from the bubble phase.

Rewriting the above equation (17), one obtains, for vigorously bubbling beds:

$$U_b = \frac{U_0 - (1 - \delta)U_{mf}}{\delta} \cong \frac{U_0 - U_{mf}}{\delta} \tag{18}$$

If the height between two successive bubbles is h, then

$$n = \frac{U_b}{h} \tag{19}$$

and

$$\delta = \frac{\frac{\pi}{6} d_b^3}{\frac{\pi}{4} d_b^2 h} = \frac{2}{3} \frac{d_b}{h} \tag{20}$$

The values of d_b, U_{br}, U_b, δ, and h can be determined from (15), (16), (17), (19), and (20), with a known value of n. The value of n can be obtained from experimental data.

On the other hand, the tendency for the bubbles to break up increases with bubble size. This limiting size is computed as follows: when the bubble breaks up, it does so in such a way that the solid particles go up through the center of the bubble and cause the separation of one cavity into two smaller cavities, as shown in Figure 5-2.

The upward velocity of gas through a large rising bubble is assumed to be nearly U_{br}. If $U_{br} > U_t$, then small entrained particles go up through the middle of the bubble and the bubble breaks up. For this condition, a new n should be introduced to make $U_{br} = U_t$.

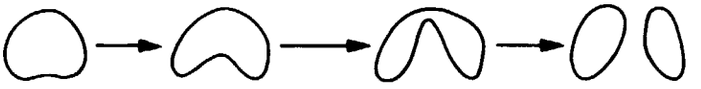

Figure 5-2. Mechanism of Bubble Breakup

SECTION VI

GAS INTERCHANGE BETWEEN BUBBLE, CLOUD, AND EMULSION

The flow pattern of gas through the fluidized bed can now be obtained from the above analysis. First, mass transfer coefficients $(K_{be})_b$, $(K_{bc})_b$, and $(K_{ce})_b$ are defined in the following way, which includes consideration of diffusion and gas circulation:

$$-\frac{1}{V_b}\frac{dN_{Ab}}{dt} = (K_{be})_b \cdot (C_{Ab} - C_{Ae})$$

$$= (K_{bc})_b \cdot (C_{Ab} - C_{Ac}) \qquad (21)$$

$$= (K_{ce})_b \cdot (C_{Ac} - C_{Ae})$$

Here V_b is the volume of bubble; N_{Ab} is the number of moles of silane in the bubble; and C_{Ab}, C_{Ac}, and C_{Ae} are the mean concentrations of silane in bubble, cloud, and emulsion, respectively. Thus $(K_{be})_b$ is the volume of gas going from bubble to emulsion divided by volume of bubbles in bed per unit time. The other two are defined similarly. From the above equations:

$$\frac{1}{(K_{be})_b} = \frac{1}{(K_{bc})_b} + \frac{1}{(K_{ce})_b} \qquad (22)$$

$(K_{bc})_b$ is now obtained in the following way. There are two modes of gas movement between the bubble and the cloud. The first is gas flow into and out of a single bubble, and the second is the mass transfer by diffusion. Assuming a Davidson's model (Reference 3-1) for bubbles, then

$$(K_{bc})_b = 4.5\frac{U_{mf}}{d_b} + 5.85\frac{D^{1/2}\, g^{1/4}}{d_b^{5/4}} \text{ and} \qquad (23)$$

$$(K_{ce})_b = 6.78\frac{\epsilon_{mf} D U_b^{1/2}}{d_b} \qquad (24)$$

where $(K_{ce})_b$ involves the diffusion process only.

SECTION VII

MASS TRANSFER RATE TO SOLID PARTICLES IN BUBBLING BED

The mass transfer rate to solid particles can now be derived using γ_b as the volume of solids dispersed in bubbles divided by volume of bubbles. The terms γ_c and γ_e are similarly defined in the Definition of Symbols. The quantity γ_b is experimentally determined.

For convenience the mass transfer process is considered to be an absorption by the solid of the silane gas present in the fluidizing gas stream. Since the flow of gas in the emulsion is very small, its minor contribution to total flow can reasonably be ignored. Thus, the fresh gas enters the bed only in the form of gas bubbles; moreover, for steady-state operations the measure of adsorption of silane is given by the decrease in its concentration within the bubbles. The mass transfer coefficient for such processes can be reported in a number of ways.

Based on total surface of particles, an overall mass transfer coefficient $(k_d)_b$ (cm/sec) can be defined as

$$-\frac{1}{\text{surface}}\frac{dn_{Ab}}{dt} = (k_d)_b\,(C_{Ab} - C_{As}) \tag{25}$$

or in terms of the falling concentration of silane in the rising bubbles:

$$-\frac{dC_{Ab}}{dt} = -U_b\frac{dC_{Ab}}{d\ell} = (k_d)_b\,\frac{\text{surface of solids}}{\text{volume of bubble}}\,(C_{Ab} - C_{As}) \tag{26}$$

$$= \frac{(k_d)_b a}{\delta}\,(C_{Ab} - C_{As}) = (K_d)_b\,(C_{Ab} - C_{As})^*$$

where C_{Ab} is the mean concentration of silane in the bubble, C_{As} is the proper concentration measure of silane at the surface of the solid, and $a = 6(1 - \epsilon_f)/d/\phi_s$.

The morphology of the silane present in a bubble as the bubble rises through the bed is considered now. Some of the silane is adsorbed by solids within the bubble; another portion is transferred to the

---

*Although expressed in bubble-related terms, this equation covers the deposition process in all phases.

cloud where a part of it is adsorbed and where the rest is transferred further into the emulsion. The silane in the emulsion is assumed to be completely adsorbed, which is reasonable in light of the long contact time in the emulsion and the rapidity of the overall adsorption process. Thus we have

$$\text{overall adsorption} = \begin{matrix}\text{adsorption in the} \\ \text{cloud by solids}\end{matrix} + \begin{matrix}\text{transfer to} \\ \text{the cloud}\end{matrix}$$

where

$$\begin{matrix}\text{transfer to} \\ \text{the cloud}\end{matrix} = \begin{matrix}\text{adsorption in the} \\ \text{cloud by solids}\end{matrix} + \begin{matrix}\text{transfer to} \\ \text{the emulsion}\end{matrix}$$

and where

$$\begin{matrix}\text{transfer to} \\ \text{the emulsion}\end{matrix} = \begin{matrix}\text{adsorption in the} \\ \text{emulsion by solids}\end{matrix}$$

In symbols, the above becomes

$$-\frac{dC_{Ab}}{dt} = -U_b \frac{dC_{Ab}}{d\ell} = (K_d)_b (C_{Ab} - C_{As}) \qquad (27)$$

$$= \gamma_b K_{d,t} a' (C_{Ab} - C_{As}) + (K_{bc})_b (C_{Ab} - C_{Ac})$$

and

$$(K_{bc})_b (C_{Ab} - C_{Ac}) = \gamma_c k_{d,mf} a' (C_{Ac} - C_{As}) + (K_{bc})_b (C_{Ac} - C_{Ae}) \qquad (28)$$

$$(K_{ce})_b (C_{Ac} - C_{Ae}) = \gamma_e K_{d,mf} a' (C_{Ae} - C_{As}) \qquad (29)$$

and

$$a' = \frac{\text{surface area}}{\text{volume of solid}} = \frac{6}{\phi_s d_p} \qquad (30)$$

By eliminating C_{Ac} and C_{Ae} from Equation (27), the general expression for the mass transfer coefficient, or Sherwood number, is obtained

$$(K_d)_b = \frac{6(1-\epsilon_f)D}{\delta\phi_s d_p^2}Sh_{overall}$$

$$= \gamma_b B_d \frac{Sh_t}{Sh_{mf}} + \cfrac{1}{\cfrac{1}{(K_{bc})_b} + \cfrac{1}{\gamma_c B_d + \cfrac{1}{\cfrac{1}{(K_{ce})_b} + \gamma_e B_d}}} \tag{31}$$

where

$$B_d = \frac{6D}{\phi_s d_p^2} Sh_{mf}(sec^{-1}) \tag{32}$$

Thus with known values for Sh_t and Sh_{mf}, and calculated gas interchange rates, mass transfer coefficients and Sherwood numbers in fluidized beds can be estimated.

$K_{d,t}$ is the mass transfer coefficient obtainable by using the single sphere diffusion formula

$$Sh_t = \frac{K_{d,t}d_p}{D} = 2.0 + 0.6\, Sc^{1/3}\, Re_{p,t}^{1/2} \tag{33}$$

where

$$Sc = \frac{\mu}{\rho D}$$

$$Re_{p,t} = \frac{d_p U_t \rho_g}{\mu}$$

SECTION VIII

PARTICLE GROWTH RATE IN A PARTICULATE BED

Based on total surface of particles, an overall mass transfer equation can be written

$$-\frac{1}{\text{surface of particles}}\frac{dN_A}{dt} = \frac{-\text{volume of bed}}{\text{surface of particles}}\frac{dC_A}{dt}$$

$$= -\frac{\phi_s d_p}{d(1-\epsilon_f)}\frac{dC_A}{dt}$$

$$= K_d(C_A - C_s)$$

Since $C_s = 0$, due to the fact that this case is diffusion-controlled (Reference 8-1), the above equation can be easily integrated. The above equation can be rewritten

$$-\frac{dC_A}{dt} = K_d' C_A$$

where

$$K_d' = K_d \cdot g(1-\epsilon_f)/\phi_s d_p$$

$$d\ln C_A = -dl$$

$$C_A = C_{Ai} \exp\ (1K_d' 1)$$

The average concentration of silane in the bed which could be used in average bed deposition calculation is

$$\bar{C}_A = \frac{C_{Ai}(1 - e^{-k_d' L_f})}{k_d' L_f} \qquad (35)$$

Therefore, the average mass transfer in the bed per unit surface of solids is given by

$$-\frac{1}{\text{surface}}\frac{dN_A}{dt} = k_d\bar{C}_A \qquad (36)$$

Finally, the particle growth rate is calculated from this result as

$$\frac{dR}{dt} = \frac{1}{\text{surface}}\frac{dN_A}{dt}\frac{Mw}{\rho_s} = k_d\bar{C}_A \qquad (37)$$

where Mw is the molecular weight of silicon, and ρ_s is its density.

SECTION IX

PARTICLE GROWTH RATE IN A BUBBLING BED

A similar procedure can be followed to obtain the particle growth rate for the bubbling bed, except that U_b, $(k_d)_b$ should be used in place of U_0, k_d. The result is that the average concentration of silane in the bubbles is

$$\bar{C}_{Ab} = \frac{U_b \cdot C_{Ai} \left(1 - e^{\frac{\left(-(k_d)_b \cdot L_f\right)}{U_b}}\right)}{(k_d)_b \cdot L_f} \tag{38}$$

The average mass transfer in the bed and the particle growth rate are given as,

$$-\frac{1}{\text{surface}} \frac{dN_{Ab}}{dt} = (k_d)_b \bar{C}_{Ab}, \tag{39}$$

$$\frac{dR}{dt} = (k_d)_b \bar{C}_{Ab} \cdot \frac{M_w}{\rho_s} \tag{40}$$

SECTION X

ENTRAINMENT

Experimental data indicate that the entrainment rate can be expressed as the entrainment coefficient k, which is given in Figure 10-1.

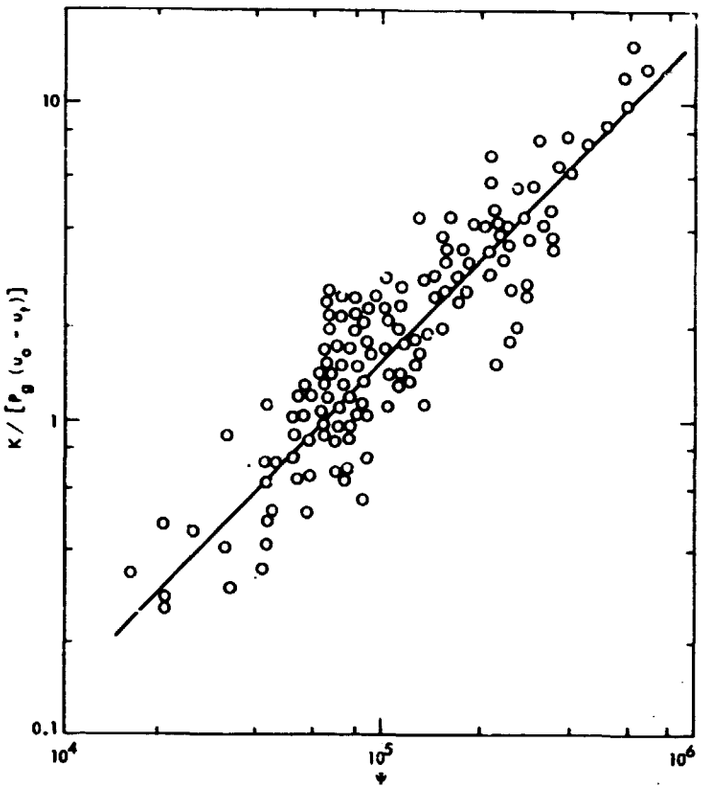

Figure 10-1. Entrainment Correlation Given (Reference 9-1):

$$\psi = \left[\frac{(u_o - u_t)^2}{g d_p}\right]^{0.5} \left(\frac{d_p u_t \rho_g}{\mu}\right)^{0.725} \left(\frac{\rho_s - \rho_g}{\rho_g}\right)^{1.15}$$

SECTION XI

OVERALL MASS BALANCE OF THE BED

The following definitions are used:

F_0 feed rate, F_1: overflow rate,

F_2 elutriation rate

P_0 particle size distribution in feed

P_1 particle size distribution in overflow

P_2 particle size distribution in elutriation

P_b particle size distribution in bed

If F_0 and P_0 are known, then there are five unknowns to be calculated and these are obtained by the following five relationships (refer to Reference 10-1):

(1) $P_1 = P_b$, for a well-mixed fluidized bed

(2) $F_1 + F_2 - F_0$ = total solid generation in bed.

(3) From the definition of k, it follows that (see Reference 10-1)

$$P_2(R) = k(R)\, WP_b\,(R)/F_2.$$

(4) By definition,

$$\int_{P_1}^{R} dR = 1.$$

(5) In unit time, the mass balance on particles of size between R and R + dR is given as follows:

$$\begin{pmatrix}\text{solids}\\ \text{entering in}\\ \text{feed}\end{pmatrix} - \begin{pmatrix}\text{solids}\\ \text{leaving in}\\ \text{overflow}\end{pmatrix} - \begin{pmatrix}\text{solids}\\ \text{leaving in}\\ \text{carryover}\end{pmatrix} + \begin{pmatrix}\text{solids growing into}\\ \text{the interval from}\\ \text{a smaller size}\end{pmatrix}$$

$$- \begin{pmatrix}\text{solids growing out}\\ \text{of the interval to}\\ \text{a larger size}\end{pmatrix} + \begin{pmatrix}\text{solids generation}\\ \text{due to growth}\\ \text{within interval}\end{pmatrix} = 0$$

This rewrites as:

$$F_0P_0(R) - F_1P_1(R) - Wk(R)P_1(R)$$

$$- W \frac{d\frac{dR}{dt}(R)\, P_1(R)}{dR} + \frac{3W}{R} P_1(R) \frac{dR}{dt}(R) = 0 \qquad (41)$$

SECTION XII

CALCULATION SAMPLES

A computer program, incorporating the mathematical model described so far, was written. A listing of the program is attached as an appendix. The properties of the gas stream and silicon particles, the composition of the gas stream, bed diameter, initial bed weight, particle size distribution of feed particles, production rate, maximum particle size in the bed, the differential particle size interval and the number of orifices in the gas distributor were the input parameters to the model. The initial value for the average particle size in the bed is assumed to be the harmonic average of the feed particle size distribution. The computer program was found to converge to final value of average bed particle size in two iterations. The mathematical modeling equations were organized in various subroutines. The main program defines the variables, arrays and functions, sets the starting values for computation, provides the calling sequence for the subroutines, and outputs the calculated data. The output of the program consists of gross fluidization characteristics, bubble characteristics, mass transfer coefficients, particle size distributions and mass flow rates of feed, outflow or entrainment and overall growth rate of particles. The minimum particle size of the feed $_{p,min.f}$ and the superficial velocity U_0 are varied in three calculations as shown in the table for simplicity. The particle size distribution of the feed was assumed to be uniform. Table 12-1 shows the results of some sample modeling calculations using this computer program.

The results show that for the given set of values of variables, the minimum fluidizing velocity U_{mf}, the terminal velocity U_t, the bed height L_f, the necessary feed rate F_0, the entrainment rate F_2, and the average particle size $\bar{d}_p$ are listed. The convergence of the computation is very fast; at the second iteration, the asymptotic solution is obtained.

F_1 was chosen after several trial and error calculations so that smooth bed operation is achieved. Such choice of numbers is delicate due to the simplistic approach taken in the modeling.

As compared to the first example, the second example shows the results when the superficial velocity is increased. This shows a smaller amount of F_0 necessary than in the first case due to more deposition from the higher flow rate. The third example is for the higher average feed particle size, showing the reduced reaction due to smaller surface area.

The results of the computer program were listed against experimental fluidization data obtained from fluidization of silicon particles with argon in a 1-in. fluidization column. The results of the comparison are shown in Table 12-2. The mode of fluidization indicated by the model is consistently wrong. It shows that the simple Froude number criterion used in the model is not sufficient to predict the mode of fluidization. The agreement between bed pressure drop data and calculated

Table 12-1. Examples of Calculational Results

- Input Parameters

d_t = 5 cm	C_{Ai} = 5.0×10^{-6} g-mole/cm^3
ρ_g = 1.23×10^{-4} g/cm^3	$r_{p,max,f}$ = 75 μ
ρ_s = 2.44 g/cm^3	$\bar{W}$ = 200 g
ρ = 3.45×10^{-4} poise	F_1 = 0.92 g/sec
D = 5.809 cm^2/sec	

- Results

	$r_{p,min,f}$ = 25 μ	25 μ	45 μ
	U_0 = 19.2 cm/sec	24.0 cm/sec	19.2 cm/sec
U_{mf}	3.78 cm/sec	4.69	5.36
U_t	9.62 cm/sec	9.62	31.2
L_f	8.5 cm	8.5	9.4
F_0	0.032 g/sec	0.023	0.048
F_2	0.011 g/sec	0.010	0.009
$\bar{d}_p$	150.1 μ	167.2	178.8

bed pressure drop is excellent. The agreement between experimental and calculated values for minimum fluidization velocity is rough. This discrepancy may be due to several reasons. The model equation used for minimum fluidization velocity is a simplified low Reynolds number approximation of the more rigorous form. The experimental data itself was prone to considerable error. In many runs (1, 2, 3, and 7) the minimum fluidization velocity was determined by visual observation of bed behaviour. In cases where the U_{mf} was determined from combined packed bed and fluidized bed data (6, 8, 9 and 10) the agreement is much better. Considering the possible significant experimental error in determining the minimum fluidization velocity, the computer model predicted the fluidized bed performance quite well.

Table 12-2. Comparison of Experimental Fluidization Data with Computer Model Results

Run No.	Particle Size, μ	Weight of Particles, g	Particle Diameter, μ	U_{mf}, cm/sec	ΔP, mm H_2O	Mode of Fluidization
1	74-104	48.2	89.00[a] 88.24[b]	0.405[c] 0.513[d] -26.6[e]	77.00[c] 76.20[d] 1.56[e]	Bubbling[c] Particulate[d]
2	74-104	79.9	89.00 88.24	0.404 0.513 -26.6	135.0 126.5 7.04	Bubbling Particulate
3	74-104	106.9	89.00 88.24	0.406 0.513 -26.6	180.0 172.1 4.38	Bubbling Particulate
6	53-74	29.0	63.50 65.74	0.33 0.28 15.15	44.00 45.84 -4.18	Bubbling Particulate
7	53-74	44.2	63.50 65.74	0.44 0.28 36.4	67.00 69.88 -4.3	Bubbling Particulate
8	74-104	30.7	89.00 88.24	0.520 0.513 1.35	46.00 48.54 -5.52	Bubbling Particulate
9	104-147	30.9	125.5 132.2	0.997 1.151 -15.45	51.00 48.84 4.24	Bubbling Particulate

Table 12-2. Comparison of Experimental Fluidization Data with Computer Model Results (Continuation 1)

Run No.	Particle Size, μ	Weight of Particles, g	Particle Diameter, μ	U_{mf}, cm/sec	ΔP, mm H_2O	Mode of Fluidization
10	104-147	45.8	125.5	1.000	78.00	Bubbling
			132.2	1.151	72.40	particulate
				-15.1	8.2	

[a]Arithmetic average of the particle sizes.
[b]Harmonic average of the particle sizes calculated by the computer model.
[c]Experimental value.
[d]Calculated value from the computer model.
[e]Percent deviation of calculated value from the experimental value.

SECTION XIII

CONCLUSIONS

As mentioned before, this modeling was intended for a simple description of the fluidized bed. Thus there are many assumptions and treatments which may not be practical. Further improvements for the model will be conducted as experimental data become available.

One such improvement area is the modeling of the fine-particle fluidized bed modeling. Although it may not be necessary if one wants to use larger-size particles when recycling some of the products by crushing them and using as the bed, it would still be beneficial to have this option of using fine particles. This modeling would be difficult at this time because very few experimental data are available. However, the JPL in-house experimental work is likely to yield the needed information for this important area of fluidized bed silicon production.

Another improvement may come from consideration of gas interchange rate at the surface of solid particles. Unlike the implicit assumption that the same number of moles of gas come out as the number of moles of gas going in, two moles of hydrogen come out for each mole of silane. This might help the fluidization of fine particles, too.

This report was reviewed by Professor T. Fitzgerald of OSU, acting as a consultant to LSSA. Dr. Fitzgerald (Reference 13-1) suggested that it might be useful to control the bubble size by artificial means: for example, vertical rods in the bed to break the bubbles larger than the size of the spacing. The reason is that, in the fluidized bed modeling, the prediction of the size of the bubble is the most difficult task (the error could be as high as 500%). This would improve the accuracy of the modeling.

He also suggested that the following assumptions may prove troublesome, and may be modified to fit real data, although these considerations will make the model complex, and the improved accuracy may or may not justify the effort for this particular task.

(1) Assumption that $C_{AS} = 0$.

(2) Assumption that P_b in bubble is same as P_{-b} in emulsion.

(3) No consideration of nucleation.

(4) Neglect of flow contribution entering emulsion phase.

This modeling is yet to be confirmed by experimental data. The experiments are underway.

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APPENDIX

Mathematical Model Program Listing

```
-FOR,IS SILICON
      REAL J
C AAIR(I), AN ARRAY, IS EQUIVALENT TO AIR(RPMIN+I*DELR-0.5*DELR), A FUNCTION.
C THIS IS INTRODUCED TO SAVE THE COMPUTER TIME BY STORING ONCE COMPUTED NUMBERS.
C ALL OTHERS ARE SIMILAR.
      COMMON AAIR(200),AABR(200),AP0(200),AP1(200),AP2(200)
      COMMON ARRR(200),AAK(200)
      COMMON UO,UMF,UT,DP,DT,H,CS,CSIL,CH2,ALMF,ALF,RHOS,RHOG,AMU
      COMMON EMF,W,F0,F1,F2,G,RE,DELP,RET,CDP,UOR,NOR,TDH,AN,DB
      COMMON DELR,UBR,UB,DELTA,UF,ALPHA,US,I,AKCEBS,UE,DSA,DSR,RC,Q
      COMMON AKDBA ,CABAV2
      COMMON SHMF
      COMMON JKL,SH1,ZETA,BD,        GAMMAE,SHT,AKDBAV,CABAV
      COMMON EF,AKBCB,AKCEB,AKBEB,D,GAMMAC,GAMMAB,ETA,SH,AKDB,SC
      COMMON REP,ZAA,ZBA,VA,AT,GROWTH,PHIS,FR,AOR,Y,CAB
      COMMON AMW,RPMIN,RPMAXF,RPMAXB,TOT
C     DT = DIAMETER OF BED
C     DP = PARTICLE DIAMETER
C     EMF = MINIMUM FLUIDIZING VOIDAGE
C     RHOG = GAS DENSITY
C     RHOS=SOLID DENSITY
C     AMU = VISCOSITY COEFF
C     PHIS=SPHERICITY
C     DELPMIN = PRESSURE LOSS ACROSS DISTRIBUTERS
C  D = DIFFUSION COEFFICIENT (CM**2/SEC)
C Y=LOGARITHMIC MEAN FRACTION OF INERT GAS
C     W=BED WEIGHT
C CAB=MEAN CONCENTRATION OF SILANE IN THE BUBBLE (GR-MOLE/CC)
C     RPMIN = MIN PARTICLE SIZE
C     RPMAXB = MAX PARTICLE SIZE IN BED
C     RPMAXF = MAX PARTICLE SIZE IN FEED
C RRR = PARTICLE GROWTH RATE IN CM/SEC
C     F1 = OUTFLOW FROM BED
C     P0 = PARTICLE SIZE DISTRIBUTION - FEEDING
C     P1 = PARTICLE SIZE DISTRIBUTION - BED AND OVERFLOW
C     P2 = PARTICLE SIZE DISTRIBUTION - ENTRAINMENT
C     F0 = MASS FLOW RATE - FEEDING
C     F1 = MASS FLOW RATE - OVERFLOW
C     F2 = MASS FLOW RATE - ENTRAINMENT
C     G = GRAVITATIONAL CONSTANT
C     ARBITRARILY ASSUME THE MAXIMUM PARTICLE SIZE IN BED.
      READ 1000,DT,PHIS,RHOG,RHOS,AMU,W,F1,EMF,Y,CAB
     *,RPMAXF,RPMAXB,RPMIN,NOR,AMW,D,G,DELR
C EVERYTHING IS EXPRESSED IN CGS UNITS! NO CONVERSION FACTOR IS THUS NEEDED.
      PRINT 888,DT,PHIS,RHOG,RHOS,AMU,W,F1,EMF,Y,CAB
     *,RPMAXF,RPMAXB,RPMIN,NOR,AMW,D,G,DELR
  888 FORMAT(1H ,///- DT=-,E10.4,- PHIS=-,E10.4,- RHOG=-,E10.4,
     1- RHOS=-,E10.4,- AMU=-,E10.4,- W=-,E10.4,/- F1=-,E10.4,- EMF=-,E10
     2.4,- Y=-,E10.4,- CAB=-,E10.4,- RPMAXF=-,E10.4,- RPMAXB=-,E10.4,/
     3- RPMIN=-,E10.4,- NOR=-,E10.4,- AMW=-,E10.4,- D=-,E10.4,- G=-
     4 ,E10.4,- DELR=-,E10.4)
      DPMIN=RPMIN*2.
      IIJ=0
    6 IIJ=IIJ+1
      IF(IIJ.EQ.1) GO TO 153
C
C PRINT COMPUTED QUANTITIES AFTER EACH CYCLE.
      RPMAX=RPMAXB
      NN=(RPMAX-RPMIN)/DELR +0.5
```

```
C IMPORTANT... RPMIN,RPMAXF,AND RPMAXB SHOULD BE MULTIPLES OF DELR
      DO 151 I=1,NN,5
      RJ=RPMIN+I*DELR-0.5*DELR
      P0J=AP0(I)
      P1J=AP1(I)
      P2J=AP2(I)
      RRRJ=ARRR(I)
      AIRJ=AAIR(I)
      ABRJ=AABR(I)
      PRINT 152,RJ,P2J,P1J,RRRJ,P0J,AIRJ,ABRJ
  152 FORMAT(1H ,-R,P2,P1,DR/DT ,P0J,AIR,ABR=-,7E10.4)
  151 CONTINUE
  153 CONTINUE
 1000 FORMAT (8E10.4)
      IF(IIJ.NE.1) GO TO 47
C     ARBITRARILY ASSUME THE MAXIMUM PARTICLE
C     SIZE IN BED MAYBE 1000 MICRONS = DPMAXB
C
C     AVERAGE PARTICLE SIZE))) INITIAL
      ARP=0.
      NN=(RPMAXF-RPMIN)/DELR+0.5
      DO 11 I=1,NN
      R =RPMIN+I*DELR-0.5*DELR
      ARP=ARP+P0(R)/R*DELR
   11 CONTINUE
      RP=1./ARP
      PRINT 55, RP
   55 FORMAT( 1H0,/- RP= -,E10.4)
      GO TO 13
C
C     AVERAGE PARTICLE SIZE))) ITERATION
   47 CONTINUE
      BRP=0.
      NN=(RPMAXB-RPMIN)/DELR+0.5
      DO 12 I=1,NN
      R =RPMIN+I*DELR-0.5*DELR
      BRP=BRP+P1(R)/R*DELR
   12 CONTINUE
      RP=1./BRP
      PRINT 55, RP
C
   13 CONTINUE
C
      DP=RP*2.
C     AT = BED AREA
      AT=DT**2*3.14159/4.
C
C     COMPUTE U0.
      CALL TESTU
C
C     DETERMINE THE TYPE OF FLUIDIZATION.
      CALL TYPE
C
C     DETERMINE THE DISTRIBUTOR PARAMETERS.
      CALL DISTRI
C
C     COMPUTE TDH
      CALL TDHS
C
```

```
C     CHARACTERIZE BUBBLES.
      CALL BUBBLE
C
C     CHARACTERIZE EMULSION PHASE.
      CALL EMULSI
C
C     CALCULATE GAS INTERCHANGE COEFFS. BETWEEN BUBBLE AND EMULSION.
      CALL GASINT
C
C     CALCULATE MASS TRANSFER COEFFS.
      CALL TRANSF
C
      IF(IIJ.NE.1) GO TO 121
      NN=(RPMAXB-RPMIN)/DELR+0.5
C
      DO 111 I=1,NN
      IF(JKL.EQ.2) GO TO 211
      ARRR(I)=RRR(RPMIN+I*DELR-0.5*DELR)
      GO TO 111
  211 ARRR(I)=RRR2(RPMIN+I*DELR-0.5*DELR)
  111 CONTINUE
      DO 112 I=1,NN
      AAK (I)=AK (RPMIN+I*DELR-0.5*DELR)
  112 CONTINUE
      DO 113 I=1,NN
      APO (I)=PO (RPMIN+I*DELR-0.5*DELR)
  113 CONTINUE
      DO 114 I=1,NN
      AAIR(I)=AIR(RPMIN+I*DELR-0.5*DELR)
  114 CONTINUE
      DO 115 I=1,NN
      AABR(I)=ABR(RPMIN+I*DELR-0.5*DELR)
  115 CONTINUE
  121 CONTINUE
C
      TOT=TOTAL(RPMAXB)
C
      DO 116 I=1,NN
      AP1 (I)=P1 (RPMIN+I*DELR-0.5*DELR)
  116 CONTINUE
C
C     BALANCE MASS FLOWS.
      CALL MASSFL
C
      DO 117 I=1,NN
      AP2 (I)=P2 (RPMIN+I*DELR-0.5*DELR)
  117 CONTINUE
C     ONLY TWO ITERATION IS ENOUGH.
      IF(IIJ.LE.3) GO TO 6
      STOP
C
      FUNCTION PO(Z3)
C  INPUT PARTICLE SIZE DISBRITUTION
      PO=0.
      IF(Z3.GE.RPMIN.AND.Z3.LE.RPMAXF) PO     =1./(RPMAXF-RPMIN)
      RETURN
C
C
      SUBROUTINE TESTU
```

```
C  THIS SUBROUTINE IS TO CALCULATE THE OPERATIONAL VELOCITY RANGE.
C     ALMF = BED HEIGHT AT MF
      ALMF=W/3.14159*4.*DT**2/(1.-EMF)/(RHOS-RHOG)/G
      DELP=W/3.14159*4./DT**2
C UNTIL WE KNOW UO,WE DON-T KNOW RE. SO THIS IS TRIAL AND ERROR METHOD.
C  1 ASSUME RE.LT.20   2  COMPUTE UMF   3  DECIDE UO(UMF,UT)  4  CALCULATE RE
C  HOWEVER, IN THIS PARTICULAR CASE UO=2.*UT
      RE = 1
      IPASS=0
C     DELP = PRESSURE LOSS ACROSS BED
C     RE = REYNOLDS NO.
C     UMF = MIN FLUIDIZING VELOCITY
C     UT = TERMINAL VELOCITY
   11 IPASS = IPASS+1
      UMF = DP**2*(RHOS-RHOG)*G/1650./AMU
      UT = G*(RHOS-RHOG)*DPMIN**2/18./AMU
      IF(RE.GE.20.) UMF = SQRT(DP*(RHOS-RHOG)*G/24.5/RHOG)
      IF(RE.GE.20.) PRINT 51
C     USE ANY UO. JUST PLACE THE DESIRED ONE IN THE LAST POSITION.
      UO=0.5*UT
      UO=.25*UT
      UO=1.5*UT
      UO=2.*UT
      UMF2=2.*UMF
      UO=2.5*UT
      UO=19.23
      IF(UO.LT.UMF2) UO=UMF2
      RE=DP*RHOG*UO/AMU
      IF(IPASS.EQ.1) GO TO 11
   51 FORMAT (1H0,- RE IS LARGER THAN 20-)
      IF(UMF.LE.UT) GO TO 32
   21 PRINT 31
   31 FORMAT(1H0,- UT IS SMALLER THAN UMF-)
   32 CONTINUE
      PRIN  61,UMF,UT,RE,ALMF,DELP,UO
   61 FORMAT(1H0,-  UMF,UT,RE,ALMF,DELP,UO=-,6E10.4)
   41 RETURN
C
C
      SUBROUTINE TYPE
C     FR = FROUDE NO
C     REPMF = RE AT MF CONDITION
C     ANUMBER = CRITERION OF TYPE OF FLUIDIZATION
C  THIS SUBROUTINE IS TO CHECK THE DEGREE OF FLUIDIZATION IN BED
      FR = UMF**2/DP/G
      REPMF = DP*UMF*RHOG/AMU
      ANUMBE=FR*REPMF*(RHOS-RHOG)/RHOG*ALMF/DT
      IF(ANUMBE.GE.100.) PRINT1
      IF (ANUMBE.LT.100.) GO TO 5
    1 FORMAT (1H0,////-  BUBBLING FLUIDIZATION-)
      UTEST = 2*UMF
      IF (UO.GE.UTEST) PRINT 2
    2 FORMAT (1H0,- THIS IS VIGOROUS-)
      IF(UO.LT.UTEST) PRINT 3
    3 FORMAT (1H0,- THIS IS NOT VIGOROUS-)
    5 CONTINUE
      IF(ANUMBE.LT.100.) PRINT 4
    4 FORMAT (1H0,- PARTICULATE FLUIDIZATION-)
      IF(ANUMBE.GE.100.) OKL=1
```

```
      IF(AMOMB.LT.100.) JKL=2
      RETURN
C
C
      SUBROUTINE DISTRI
C  THIS SUBROUTINE IS TO CALCULATE THE DISTRIBUTOR
C     VELRAT = VELOCITY RATIO
C     AOR = ORIFICE AREA
C     CDP = ORIFICE COEFFICIENT
C     NOR = NUMBER OF ORIFICE
C     UOR = ORIFICE VELOCITY
      DELP1 = DELP*0.1
      DELP2=3.5E+04
C     DELP2 IS EQUIVALENT TO 35 CM H2O
C     REFER TO P67 OF KUNII, EQ 34. THE THIRD TERM
C     IS NEGELECTED HERE
      DELPD=DELP1
      IF (DELP2.GE.DELP1) DELPD=DELP2
      IF(DELP2.GT.DELP1) PRINT 11
   11 FORMAT(1H0,///- WE CAN INCREASE WEIGHT OF THE BED WITHOUT PENALIZING THE
     *DESIGN DISTRIBUTOR PRESSURE LOSS-)
      RET = DT*RHOG*UO/AMU
C  REFER TO FIG 12 OF P88
      IF(RET.GE.30.) CDP=0.6
      IF(RET.LT.30.) CDP=0.4*(ALOG(RET)-ALOG(2.))/(ALOG(30.)-ALOG(2.))
     *+0.2
      UOR = CDP* SQRT(2.      *DELPD/RHOG)
      VELRAT=UO/UOR
      AOR = VELRAT/NOR
      PRINT 21,RET,CDP,UOR,VELRAT,AOR
   21 FORMAT(1H0,-RET,CDP,UOR,VELRAT,AOR=-,5E10.4)
      RETURN
C
C
      SUBROUTINE TDHS
C  THIS SUBROUTINE IS TO CALCULATE TDH
C     REFER TO P94
C     TDH = TRANSPORT DISENGAGING HEIGHT
C     DUMMY SUBROUTINE
      RETURN
C
C
      SUBROUTINE BUBBLE
C     UF = UPWARD VELOCITY OF GAS IN EMULSION AT MF
C     DELTA = VOLUME FRACTION OF BUBBLES
C     RB = RADIUS OF BUBBLE
C  THIS SUBROUTINE COMPUTES BUBBLE PROPERTIES.
C     REFER TO CHAP 4
C     DB = DIAMETER OF BUBBLE
C     AN   BUBBLE FREQUENCY
C     UBR = BUBBLE RISING VELOCITY
C     UB = ABSOLUTE RISE VELOCITY OF BUBBLES
C     H = HEIGHT BETWEEN TWO SUCCESSIVE BUBBLES
C     SC=SCHMIDT NO.
C     FOR SIMPLICITY OF THE CALCULATION, BASED ON THE INFORMATION IN
C     FIG 20,P129( LET-S ASSUME THAT THE BUBBLE FREQUENCY IS APPROXIMATELY
C     4 REGARDLESS OF THE SIZE OF THE BED FOR THE MOST PART OF THE BED.
      AN = 4.
      DB=1.5/AN*(UO-UMF)
```

```
      PRINT 1,DB,AN
    1 FORMAT (1H0,//-  BUBBLE SIZE, FREQUENCY =-,2E10.4)
      UBR = 22.26 * DB** (0.5)
C THE BUBBLE SIZE CANNOT BE LARGER THAN MAX STABLE SIZE. EQ.9.P121
      IF( UBR.GE.UT) UBR=UT
      DB=UBR**2/22.26**2
      RB=DB/2.
      UB=UO-UMF+UBR
      H = UB/AN
      UF = UMF/EMF
      DELTA=2./3.*DB/H
      PRINT 2,UBR,UB,H,UF,DELTA,DB
    2 FORMAT(1H0,/- UBR,UB,H,UF,DELTA, B=-,6E10.4)
      RETURN
C
C
      SUBROUTINE EMULSI
C  EMULSION PHASE CHARACTERISTICS
C     REFER TO CHAP 4 AND 5
C     RC = RADIUS OF CLOUD
C     Q = FLOW RATE OF GAS INTO AND OUT OF A BUBBLE
C     (P115)
C     ALF = HEIGHT OF BUBBLING BED (P131)
C     EF = VOID FRACTION IN BUBBLING BED(P131)
C     UE = UPWARD VELOCITY OF GAS IN EMULSION      (P152)
C     ALPHA = RATIO OF WAKE VOLUME TO BUBBLE VOLUME
C     US = MEAN DOWNWARD VELOCITY OF SOLIDS IN EMULSION
C     AKCEBS(P157) = COEFF OF SOLID INTERCHANGE BETWEEN
C     CLOUD-WAKE REGION AND EMULSION PHASE
C     DSA = AXIAL DISPERSION COEFF OF SOLIDS
C     DSR = RADIAL DISPERSION COEFF OF SOLIDS
      RC = 0.5*DB*((UBR+2.*UF)/(UBR-UF))**0.3333
C     DBMAX WOULD NOT BE DEFINED (P122 FIG.14)
      Q = 3.*UF*EMF*3.14*DB**2 * 0.25
      ALF = ALMF / (1.-DELTA)
      EF=1.-(1.-DELTA)*(1.-EMF)
      PRINT 1, RC, Q, ALF, EF
    1 FORMAT (1H0,/- RC,Q,ALF,EF,=-,4E10.4)
C FOR SIMPLICITY,BASED ON FIG.9,P150, LET-S ASSUME THAT ALPHA IS CONSTANT
      ALPHA=0.4
      US = ALPHA*DELTA*UO/(1.-DELTA-DELTA*ALPHA)
      UE=UF-US
      J = ALPHA*DELTA*UB*RHOS*(1.-EMF)
      AKCEBS = 3.*(1.-EMF)*UMF/(1.-DELTA)/EMF/DB
      DSA=ALPHA**2*EMF*DB*(UO-UMF)**2/3./DELTA/UMF
      DSR = 3./16.*DELTA/(1.-DELTA)*UMF*DB/EMF
      PRINT 2, ALPHA,US,J,AKCEBS,UE,DSA,DSR
    2 FORMAT(1H0,/- ALPHA,US,J,AKCEBS,UE,DSA,DSR=-,7E10.4)
      RETURN
C
C
      SUBROUTINE GASINT
C  GAS INTERCHANGE BETWEEN BUBBLE AND EMULISON.
C  WILL BE APPLIED TO THE CASE OF VIGOROUS
C  BUBBLING BEDS  EITHER UO,GE,2UMF OR
C  UB.GE.5UF
C  AKBCB = INTERCHANGE COEFFICIENT BETWEEN
C  BUBBLE AND CLOUD
C  AKCEB = BETWEEN CLOUD AND EMULSION
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C  AKBEB = BETWEEN BUBBLE AND EMULSION
C  THESE COEFFS. ARE ASSUMED TO BE INDEPENDENT OF INDIVIDUAL PARTICLE SIZE
      AKBCB=4.5*UMF/DB+5.85*(D**2*G/DB**5)**(0.25)
      AKCEB = 6.78 * (EMF*D*UB/DB**3)**(0.5)
      AKBEB=1./(1./AKBCB+1./AKCEB)
      PRINT 1, AKBCB,AKCEB,AKBEB
    1 FORMAT(1H0,/- AKBCB,AKCEB,AKBEB=-,3E10.4)
      RETURN
C
C
      SUBROUTINE TRANSF
C OBTAIN DIFFUSION COEFFICIENT ON TO SILICON SOLIDS
      ETA=1.
C EFFECTIVENESS FACTOR
C ETA IS TAKEN TO BE 1. SINCE THE PROCESS IS DIFFUSION CONTROLLED
C     AND THUS THE SILANE CONCENTRATION ON SOLID PARTICLES,CAS, IS NOT
C     IMPORTANT IN OUR CASE.
C     GAMMAB= VOLUME FRACTION OF SOLIDS IN BUBBLE
C GAMMAB IS ARBITRARILY CHOSEN(P202)
      GAMMAB=0.003
C     GAMMAC= VOLUME FRACTION OF SOLIDS IN CLOUD
      GAMMAC=(1.-EMF)*(3.*UMF/EMF/(UBR-UMF-EMF)+ALPHA)
C     GAMMAE= VOLUME FRACTION OF SOLIDS IN EMULSION
      GAMMAE=(1.-EMF)*(1.-DELTA)/DELTA-(GAMMAB+GAMMAC)
C     SC    = SCHMIDT NO.
      SC=AMU/RHOG/D
C     RETT  = REYNOLDS NO. WITH UT
      RETT=DP*UT*RHOG/AMU
C     REMF  = REYNOLDS NO. WITH UMF
      REMF= DP*UMF*RHOG/AMU
C     ZETA  = A VARIABLE INTRODUCED IN NELSON & GALLOWAY, WITH UO
      ZETA=(1./(1.-EF)**(1./3.)-1.)*0.3*RE**(0.5)*SC**(1./3.)
C     ZEMF  = SAME VARIABLE WITH UMF
      ZEMF=(1./(1.-EF)**(1./3.)-1.)*0.3*REMF**(0.5)*SC**(1./3.)
C     SHMF  = SHERWOOD NO. WITH MF CONDITIONS. TO BE USED IN EMULSION
C      PHASE IN BUBBLING BED
      SHMF=(2.*ZEMF+(2.*ZEMF**2*(1.-EF)**(1./3.)/(1.-(1.-EF)**(1./3.))
     +**2-2.)*TANH(ZEMF))/(ZEMF/(1.-(1.-EF)**(1./3.))-TANH(ZEMF))
C     SH1   = SHERWOOD NO. WITH UO. TO BE USED IN PARTICULATE BED.
      SH1 =(2.*ZETA+(2.*ZETA**2*(1.-EF)**(1./3.)/(1.-(1.-EF)**(1./3.))
     +**2-2.)*TANH(ZETA))/(ZETA/(1.-(1.-EF)**(1./3.))-TANH(ZETA))
C     SHT   = SHERWOOD NO. WITH UT. TO BE USED FOR SOLIDS IN BUBBLES.
      SHT=2.0+0.6*SC**(1./3.)*SQRT(RETT)
C     AKDBA2= AVERAGE MASS TRANSFER COEFF. FOR PARTICULATE BED.
      AKDBA2=SH1*D/DP
C     CABAV2=AVERAGE CONCENTRATION OF SILANE IN PARTICULATE BED.
      CABAV2=UO*CAB*(1.-EXP(-AKDBA2*ALF/UO))/AKDBA2/ALF
C     COMPUTE AVERAGE SILANE CONCENTRATION IN BED. REFER TO MY WRITEUP.
C     DP IS THE AVERAGE PARTICLE SIZE.
C     BD    = AN INTERMEDIATE VARIABLE.
      BD=6.*D*SHMF/PHIS/DP**2
C     AKDBAV= AVERAGE MASS TRANSFER COEFF. FOR BUBBLING BED.
      AKDBAV=GAMMAB*BD*SHT/SHMF+1./(1./AKBCB+1./(GAMMAC*BD+1.
     +/(1./AKCEB+1./GAMMAE/BD)))
C     CABAV =AVERAGE CONCENTRATION OF SILANE IN BUBBLING BED.
      CABAV=UB*CAB*(1.-EXP(-AKDBAV*ALF/UB))/AKDBAV/ALF
      PRINT 5,CABAV,CABAV2
    5 FORMAT(1H0,- CABAV,CABAV2=-,2E10.4)
      RETURN
```

```
C
      FUNCTION AIR(A)
C     AIR = INTEGRAL EQ.32. P339
C TO COMPUTE THE INTEGRAL I(R,RI)
      NN=(A-RPMIN)/DELR+0.5
      AIRLOG=0.
      DO 1 I=1,NN
      R9=RPMIN+I*DELR-0.5*DELR
      AIRLOG=AIRLOG+(F1/W+AAK(I))/ARRR(I)*DELR
    1 CONTINUE
      AIR=EXP(-AIRLOG)
      RETURN
C
C
      FUNCTION ABR(Z1)
C  COMPUTE AN INTERGRAL
C     ABR = INTEGRAL SHOWN IN EQ.36. P340
      ABR=0.
      NN=(Z1-RPMIN)/DELR+0.5
      DO 51 I=1,NN
      RI=RPMIN+I*DELR-0.5*DELR
      ABR=ABR+APO(I)/RI**3/AAIR(I)*DELR
   51 CONTINUE
      RETURN
C
      FUNCTION TOTAL(Z4)
C  COMPUTE AN INTEGRAL W/FO (EQ.37,P340)
      TOTAL=0.
      NN=(Z4-RPMIN)/DELR+0.5
      DO 1 I =1,NN
      R6=RPMIN+I*DELR-0.5*DELR
      TOTAL=TOTAL+R8**3/ARRR(I)*AAIR(I)*DELR*AABR(I)
    1 CONTINUE
      RETURN
C
C
      SUBROUTINE MASSFL
C     DELR = PARTICLE SIZE INCREMENT USED FOR
C     COMPUTING THE INTEGRALS
      FO=W/TOT
C  COMPUTE FO,F2,P1,P2
C  GROWTH = PARTICLE GROWTH RATE IN BED(EQ.23,P337)
      GROWTH=0
      NN=(RPMAXB-RPMIN)/DELR+0.5
      DO 1 I=1,NN
      R6=RPMIN+I*DELR-0.5*DELR
      GROWTH=GROWTH+DELR*3.*W*AP1(I)*ARRR(I)/R6
    1 CONTINUE
      F2 = GROWTH - F1+FO
      PRINT 2, FO,F1,GROWTH,F2
    2 FORMAT(1H0,//- FO,F1,GROWTH,F2=-,4E10.4)
      RETURN
C
C
      FUNCTION RRR(X1)
C     TO COMPUTE DR/DT FOR BUBBLING BED
      X=2.*X1
      RRR=(DELTA*X*PHIS/6./(1.-EF))*(CABAV*AMW/RHOS)*(GAMMAB*6.*D*SHT/
     +PHIS/X**2+1./(1./AKECB+1./(GAMMAC*6.*D*SHMF /PHIS/X**2+1./
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```
     *(1.+AKCEB*(1.+(GAMMAE*6.*D*SHMF/PI*S/X**2)))))
      RETURN
C
      FUNCTION RRR2(X)
C     TO COMPUTE DR/DT FOR PARTICULATE BED
      RRR2=SH1*D*AMW*CASAV2/RHOG/2./X
      RETURN
C
      FUNCTION AK(A)
C  THIS ROUTINE COMPUTES THE ELUTRIATION COEFFICIENT
C  FROM TRANSFE RRR=ZAA/ZP+ZBA*DP.
C     KSI - REFER TO P316. FIG 13
C     AK = ELUTRIATION CONSTANT AS A FUNCTION OF RADIUS
C     KSI = (UG-UT)/SQRT(G)*(UT*RHOG/AMU)**0.725
C     *((RHOS-RHOG)/RHOG)**1.15 * DP**0.225
C     = VA*DP**0.225
C     NO ELUTRIATION IF UG.LE.UT
      IF(UG.LE.UT) GO TO 1
      VA =(UG-UT)/SQRT(G)*(UT*RHOG/AMU)**0.725
     **((RHOS-RHOG)/RHOG)**1.15
      AK    =EXP(ALOG(0.15)+(ALOG(17.)-ALOG(0.15))*( ALOG(VA*12.*A)**
     $0.225)  -ALOG(10000.))/(ALOG(10000000.)-ALOG(10000.)))*RHOG
     *     *(UG-UT)*AT/W
      GO TO 2
    1 CONTINUE
      AK=0
    2 CONTINUE
      RETURN
C
C
      FUNCTION P1(Z6)
C     EQ.35,P340
      P1=Z6**3/ARRR(I)*AAIR(I)*AABR(I)/TOT
      RETURN
C
C
      FUNCTION P2(Z10)
C     EQ.7, P329
      P2=AAK(I)*W/F2*AP1(I)
      RETURN
      END
-MAP
-XQT
 .5   E+01 .86  E+00 .1236E-03 .244 E+01 .3454E-03 .196 E+06 .9   E+03 .5   E+00
 .4   E+00 .5   E-05 .75  E-02 .9   E-01 .45  E-02 .1   E+03 .2809E+02 .5809E+02
 .98  E+03 .5   E-03
-FIN
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