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THE EFFECT OF INTERPARTICLE FORCES ON THE STABILITY OF GAS-FLUIDIZED BEDS—I. EXPERIMENTAL EVIDENCE

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Abstract—The stability theory for homogeneous fluidized beds presented earlier is reviewed. This theory is based on the concept of the elasticity of the bed structure as a consequence of interparticle forces. It is shown that this theory explains the effect of gas viscosity and gravity. It is further shown that the elasticity modulus is increased by gas adsorption to the solid surface at elevated pressure and, thus, explains the effect of gas pressure on bed expansion. The theory is compared with experimental results obtained with fluidization of fresh cracking catalyst and polypropylene by different gases and at gas pressures up to 15 bar. It is further shown that the elasticity modulus can be used to correlate bed expansion and bubble size during heterogeneous fluidization. The stability theory of Foscolo and Gibilaro is criticized and rejected on the basis of serious mistakes made in their theoretical derivation.

INTRODUCTION

The original stability theories (Jackson, 1963; Pigford and Baron, 1965) were of a purely hydrodynamic conception of the fluidized bed and assumed that during fluidization the particles are free floating, and hence denied the possibility of interparticle forces. The outcome of these theories was that the fluidized bed would always be unstable as soon as the superficial gas velocity exceeded the incipient fluidization velocity. In the years around 1975 we published a series of papers on the effect of interparticle forces on the stability of a homogeneous gas-fluidized bed of fine solids (Rietema and Mutsers, 1973, 1975; Mutsers and Rietema, 1977a, b, 1978). These papers were a further elaboration of a short communication by Rietema (1973).

The basic idea was that as a consequence of interparticle forces an elasticity term should be introduced in the momentum equation of the solids. This elasticity term was proposed to be $E(\partial\epsilon/\partial h)$, where E is the elasticity modulus of the bed (measured in N/m^2). It is the mathematical representation of forces which result in the occurrence of dynamic waves in the particle bed. As long as the dynamic wave velocity exceeded the velocity with which a disturbance in the fluidized powder bed moves up the disturbance would be damped out and the bed would remain stable (Wallis, 1969). This theory led to a mathematical criterion which indicated that the bed will be stable when

$$\frac{\rho_p^3 d_p^4 g^2}{\mu^2 E} \leq \left[\frac{150(1 - \epsilon)}{\epsilon^2(3 - 2\epsilon)} \right]^2 \quad (1)$$

At the bubble-point velocity when $\epsilon = \epsilon_{mb}$ the equality would hold. This theory was received with much scepticism (Lockett and Gunnarsson, 1973; King and Harrison, 1982; Clift, 1985; Foscolo *et al.*, 1985),

Fig. 1. Electrical conductivity and pressure drop of charcoal bed vs superficial gas velocity.

mainly because most chemical engineers believe that such a strong influence of interparticle forces seems rather unacceptable. A remarkable formulation of this criticism is found in Foscolo *et al.* (1985): “interparticle forces emerged as the shroud under which failures to match theory[†] with experimental truth could conveniently be obscured, thus adopting a role more therapeutic than scientific”.

Hence, researchers on fluidization kept seeking a criterion based solely on hydrodynamics (Verloop and Heertjes, 1970; Oltrogge, 1972). The latest attempt in this direction is that of Foscolo and Gibilaro (1984), some 6 years ago.

EXPERIMENTAL EVIDENCE OF INTERPARTICLE FORCES

Electrical conductivity of a fluidized bed

Contrary to the assumption of free floating it was found by us (Rietema, 1967) and others (Graham and Harvey, 1965) that when a bed of fine charcoal par-

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[†]Hydrodynamic theories (Jackson, 1963; Pigford and Baron, 1965) which do not account for interparticle forces.

ticles is fluidized and two vertical electrodes are submerged in the bed the bed has a considerable electrical conductivity during all the fluidization stages (Fig. 1). This is only possible when the particles stay in contact with each other during fluidization and hence can act as attraction and friction forces on each other.

It has been suggested that this conductivity could be also caused by frequent collisions between otherwise free floating particles. A simple analysis (see Appendix 1) shows that in this way the conductivity would be a factor of 10^6 too small.

The tilting bed

That interparticle forces are real in a gas-fluidized bed is most clearly demonstrated by the tilting bed (Fig. 2) (Rietema and Mutsers, 1973). When a homogeneously fluidized bed is tilted over a horizontal axis it remains stable until a critical tilting angle α_c is reached at which the bed surface suddenly shears off. This critical tilting angle depends on the degree of bed expansion and, of course, also on particle size and particle density (Fig. 3).

This experiment clearly demonstrates that the homogeneously fluidized bed maintains a mechanical structure of a certain strength, caused by a network of interparticle contacts. The greater is the bed expansion the weaker is the mechanical structure.

The surpressure at incipient fluidization

At full fluidization the pressure drop over the bed is equal to W/A , where W is the weight of the bed, and A is the cross-sectional area of the bed. At increasing gas velocity this pressure drop is first reached at incipient

fluidization. Just beyond incipient fluidization the pressure drop increases even above this value, the difference being called the surpressure. At the same time the bed is prevented from expanding and is kept in the packed state. This is caused by the forces with which the particles act upon each other and on the wall of the bed container (Fig. 4).

With further increase in the gas velocity a point is reached where these forces are not sufficient anymore, where the bed will suddenly expand while at the same time the surpressure is strongly reduced.

STABILITY THEORY

From the theory of Wallis (1969) it is clear that the stability of a homogeneously fluidized bed is only possible if that bed has an intrinsic elasticity of sufficient strength to damp out possible perturbations that may arise from the outside in the otherwise uniform structure of the bed.

This means that due to that elasticity a force is generated by the bed structure which is proportional to the concentration gradient of the dispersed particles and directed opposite to this gradient. Hence, this force tries to restore the original structure of the bed when a perturbation passes. In a fluidized bed disturbances of the uniform concentration distribution will always move up and are called continuity waves (Wallis, 1969). Their propagation velocity V_c is given by the equation

$$V_c = (1 - \varepsilon) \frac{\partial u_o}{\partial \varepsilon} = u_o \left(\frac{3 - 2\varepsilon}{\varepsilon} \right). \quad (2)$$

Fig. 2. Tilting bed apparatus, just before (a) and just after (b) shearing off.

When such a continuity wave passes in a bed with an intrinsic elasticity the elastic force generates dynamic (elastic) waves with a propagation velocity of

$$V_d = \left(\frac{E}{\rho_p} \right)^{0.5} \quad (3)$$

where E is the elasticity modulus of the elastic medium.

When the elastic force is strong enough V_d will be larger than V_c and the disturbance will be damped out. When $V_d < V_c$ the disturbance will grow and the bed become unstable. The maximum stable bed expansion, therefore, is found when $V_d = V_c$. Using this result gives, for the elasticity modulus E :

$$E = \rho_p \left[\frac{u_o(3 - 2\epsilon)}{\epsilon} \right]^2 \quad (4)$$

With the relationship

$$u_o = \frac{(\rho_p - \rho_g)gd_p^2}{150\mu} \frac{\epsilon^3}{1 - \epsilon} \quad (5)^\dagger$$

the stability criterion can be further elaborated:

$$\text{stability if } N_r = \frac{\rho_p(\rho_p - \rho_g)^2 g^2 d_p^4}{\mu^2 E} \leq \left[\frac{150(1 - \epsilon)}{\epsilon^2(3 - 2\epsilon)} \right]^2 \quad (6)^\dagger$$

The problem is, of course, what is the origin of the elasticity. According to the theory of Rietema and Mutsers this origin is to be found in the interparticle forces which in turn have their origin in the well-known van der Waals forces. These were first quantified by London (1937) and afterwards by Lennard-Jones (1937). According to the latter the potential between two interacting molecules is given by

$$V_{ss} = C_{ss} \left(-\frac{1}{r^6} + \frac{r_{ss}^6}{2r^{12}} \right) \quad (7)$$

where r_{ss} is a characteristic parameter of the molecules (in m) and C_{ss} is the London-van der Waals constant (in Jm⁶), while r is the distance between the two molecules. When this potential is integrated over all molecules of two interacting particles [applying the integration method of Hamaker (1937)] we get the total molecular energy of the system:

$$U_m = \frac{A_{ss}d}{12z} \left[-1 + \frac{1}{420} \left(\frac{r_{ss}}{z} \right)^6 \right] \quad (8)$$

where A_{ss} is the Hamaker constant, $d = d_1 d_2 / (d_1 + d_2)$, where d_1 and d_2 are the diameters of the interacting particles, and z is the shortest distance between them. By differentiating eq. (8) with respect to z the interaction force F_i is found. In dimensionless notation with $\zeta = z/r_{ss}$

$$\phi = \frac{12F_i r_{ss}^2}{A_{ss}d} = \frac{1}{\zeta^2} \left(1 - \frac{1}{60\zeta^6} \right). \quad (9)$$

For more details one is referred to Part II. ϕ is shown as a function of ζ in Fig. 5 from which it is clear that there is an elastic range where ϕ and hence F_i increase as the distance between the particles increases.

When the distance exceeds, however, the value z_m (corresponding to ζ_m) the external force will cause the contact to be broken. The problem is even more

Fig. 3. Critical tilting angle and bed expansion vs superficial gas velocity for a fluidized bed of fresh cracking catalyst. Gases: (Δ) air, (+) H₂.

Fig. 4. Pressure drop and surpressure over a fluidized bed of fresh cracking catalyst.

Fig. 5. Dimensionless interaction force as a function of the dimensionless particle-particle distance.

[†]In the derivation of the original bubble point criterion the gas density was neglected. When this is no longer allowed the equations should be modified as indicated in eqs (5) and (6).

complicated when the interacting particles are not rigid but elastic themselves, and hence can be deformed. Further complication is caused by gas adsorption [see Cottaar and Rietema (1986)]. These theoretical considerations are all the subject of Part II.

On the basis of the above it is justified to put in a first approximation for an expanded powder:

$$F_{e,h} = E(\varepsilon_h - \varepsilon_o) \quad (10)$$

where $F_{e,h}$ is the elastic force per unit cross section in the powder at height h , and ε_h is the porosity at the same height, while ε_o is a reference porosity, and E is the elasticity modulus.

At the height $h + \Delta h$:

$$F_{e,h+\Delta h} = E(\varepsilon_{h+\Delta h} - \varepsilon_o)$$

and, hence, it is found that per unit volume of the dispersed system the net force is $E(\partial\varepsilon/\partial h)$ and thus satisfies the requirement of Wallis (1969) that this force should be proportional to the concentration gradient of the solids and directed opposite to this gradient.

It must be stressed that the elasticity modulus E is not a constant but depends on the porosity. It is determined by the forces at the contact points between the solid particles and the number of contact points per particle (generally called the coordination number). This coordination number decreases with increasing porosity (see Part III).

In a small range of the porosity, however, E will not change as long as the elasticity yield stress is not exceeded and contact points are not broken. Hence, in such a range eq. (10) holds and shows indeed that on increasing the porosity within this range the elastic force $F_{e,h}$ will increase also. This means that when slowly increasing the fluidization gas velocity the local porosity will increase in small steps (e.g. $\Delta\varepsilon = 0.002$) while during each step E will remain constant. At the end of each step one or more local contacts between particles will be broken and the local bed structure will be rearranged.

Of course, this will not happen anywhere at the same time but such a rearrangement will be spread at random over the total bed and in time. This results in a nearly continuous increase in the total bed height.

In the mathematical analysis of the problem the elastic force was introduced in the momentum equation of the dispersed solid phase which is now

$$(1 - \varepsilon)\rho_p \frac{D}{Dt} v_d + (1 - \varepsilon) \frac{\partial p}{\partial h} - E \frac{\partial \varepsilon}{\partial h} + (1 - \varepsilon)\rho_p g + F = 0 \quad (11)$$

where v_d is the velocity of the solid phase, and F is the slip force with which the dispersed phase acts on the continuous gas phase. $F = (v_d - v_c)/m$, where v_c is the linear gas velocity, and m is the so-called mobility of the solid phase. Together with the momentum equation of the gas phase and the two continuity equations (one for the gas phase and one for the solid phase) and

applying perturbation theory the critical circumstances under which a perturbation is damped out could be derived (Rietema and Mutsers, 1973; Mutsers and Rietema, 1977a) and, indeed, resulted in the already mentioned stability criterion. At the maximum stable bed expansion with porosity ε_{mb} (just no bubbles) the following equality holds:

$$E_{mb} = \rho_p \left[\frac{u_{mb}(3 - 2\varepsilon_{mb})}{\varepsilon_{mb}} \right]^2. \quad (12)$$

Measuring ε_{mb} and u_{mb} , therefore, makes it possible to determine at this porosity the elasticity modulus E_{mb} .

When the same particle system is fluidized with a gas with a higher viscosity a higher maximum stable bed expansion can be reached and thus by fluidizing with different gases with different viscosities the dependence of E on the porosity can be determined. In this way it was found (see Fig. 12) that this dependence could be well described by

$$E = E_o e^{-b(\varepsilon - \varepsilon_o)} \quad (13)$$

where E_o is the value of the elasticity modulus E at porosity ε_o , and b is a numerical constant.

Foscolo and Gibilaro (1984) also recognized the necessity of elastic waves for stability but they reject the influence of interparticle forces. Instead, they seek the origin of elastic waves in hydrodynamics. This is remarkable since from the momentum eq. (11) of the solids, but without the interparticle forces, no elastic force can be derived.

They did a mental experiment with a movable frictionless piston as the gas distributor. When this piston is moved up by means of an outward force the bed at the bottom is compressed and subsequently this compression wave moves up through the bed. They stated that this is an elastic wave with a propagation velocity of

$$V_d = \left(\frac{\partial p}{\partial \rho} \right)^{0.5}. \quad (14)$$

They also assumed that to ensure stability of the fluidized bed V_d must be larger than the propagation velocity V_c of the continuity waves. In their elaboration of the elastic wave velocity V_d they assumed that the hydrodynamic force imparted to the particles is dependent on the concentration gradient. For the relationship between the superficial gas velocity and the voidage in the bed they used the equation of Richardson and Zaki (1954):

$$u_o = u_t \varepsilon^n.$$

Finally they arrived at the following criterion:

stability if

$$\frac{(gd_p)^{0.5}}{u_t} \left(\frac{\rho_p - \rho_g}{\rho_p} \right)^{0.5} \geq 0.56n(1 - \varepsilon)^{0.5} \varepsilon^{n-1}. \quad (15)$$

With the Archimedes number

$$Ar = \frac{(\rho_p - \rho_g)\rho_p g d_p^3}{\mu^2}$$

and for $Re_t < 0.2$ this criterion can also be formulated as

$$Ar \leq \frac{45}{(1 - \varepsilon)\varepsilon^{7.6}}$$

The statement of Foscolo and Gibilaro that the compression wave in their mental experiment is an elastic wave, however, is not correct. It is just a perturbation induced by an outward force and not the consequence of an intrinsic property of the system. In contradiction to elastic waves this compression wave does not move in two directions (up and down) but only upwards. Hence, it just is a continuity wave.

Foscolo and Gibilaro also fail to indicate what might be the stabilizing force which should be proportional to the concentration gradient but directed opposite to it.

Finally the expression for the elastic wave velocity V_d [eq. (14)] which they applied does not hold for a dispersed two-phase system such as a fluidized bed but only for a homogeneous compressible one-phase system.

The theory of Foscolo and Gibilaro resembles in many aspects the theory of Oltrogge (1972) and that of Verloop and Heertjes (1970). These authors developed theories which are basically also of a pure hydrodynamic nature. Hence, their results can be presented also in the general form

$$Ar \leq f(\varepsilon)$$

where only the dependence on the porosity differs from author to author. A criticism of these theories has already been given in Rietema (1973) and will not be repeated here.

In a more recent paper Foscolo and Gibilaro (1987) present a seemingly more exact derivation of their stability criterion. In Appendix 2, we show that in this derivation they neglected a minus sign in their expression of the elastic wave velocity V_d . Elaborating correctly the hypothesis of a hydrodynamic elasticity as proposed by Foscolo and Gibilaro the final result would be instability of the powder bed at all porosities.

THE EFFECT OF GRAVITY

To test their theory Mutsers and Rietema carried out experiments with a fluidized bed mounted in a centrifugal field (Mutsers and Rietema, 1977b, 1978). For that purpose they made use of a so-called "human centrifuge" at the National Aerospace Medical Center, Soesterberg, Netherlands. This apparatus was originally constructed for the testing and training of air pilots to cope with variations in the apparent gravity.

Two powders were investigated, spent cracking catalyst and polypropylene, while both hydrogen and nitrogen were used as fluidization gases. The apparent gravity was varied by a factor 3. The fluidized bed was mounted in the cabin of the human centrifuge. This cabin was rotated on a radial arm of about 2.5 m. A

videocamera was also mounted in the cabin to observe the fluidization behaviour of the powders. For more details, see Mutsers and Rietema (1977b). At each set of conditions the bubble point porosity ε_{mb} was determined. To compare the results with the theory the elasticity modulus for each powder was assumed to be a function only of the porosity.

In Figs 6 and 7 the experimental results are plotted vs g_c/g , where g_c is the apparent gravity constant. The theoretical relation is shown in Figs 6 and 7 as well.

Gibilaro *et al.* (1986) also used the results of Mutsers and Rietema to test their theory. In Figs 6 and 7 their theoretical predictions are also given. For the spent cracking catalyst–nitrogen system their theory shows a good agreement with the experiments. For the polypropylene–nitrogen system, however, the agreement is rather poor and their theory gives a far too strong effect of gravity. For the two systems with hydrogen as the fluidization gas the discrepancy with the theory of Foscolo and Gibilaro is really very serious and strongly suggests that this theory is wrong. As a whole the theory of Rietema and Mutsers fits the experiments much better.

Fig. 6. Bubble point porosity ε_{mb} vs variation of apparent gravitational constant as measured for spent cracking catalyst fluidized in a centrifugal field with nitrogen and hydrogen, respectively. Gases: (Δ) N_2 , ($\blacktriangle$) H_2 . (—) Theoretical line, Rietema and Mutsers; (---) theoretical line, Foscolo and Gibilaro.

Fig. 7. Bubble point porosity ε_{mb} vs variation of apparent gravitational constant as measured for polypropylene fluidized in a centrifugal field with nitrogen and hydrogen, respectively. Gases: (Δ) N_2 , ($\blacktriangle$) H_2 . (—) Theoretical line, Rietema and Mutsers; (---) theoretical line, Foscolo and Gibilaro.

THE EFFECT OF GAS PRESSURE

It has been found by several authors (King and Harrison, 1982; Guedes de Carvalho and Harrison, 1975; Crowther and Whitehead, 1978) that the maximum homogeneously fluidized bed expansion increases considerably with increasing gas pressure. This was confirmed in our laboratory at Eindhoven by experiments carried out by Piepers *et al.* (1984).

The increase in bed expansion also depends on the type of fluidization gas used; with hydrogen as the fluidization gas this increase was hardly perceptible (see Fig. 8) and this suggests that the phenomenon is related in one way or another with the gas density. As remarked by King and Harrison (1982) it seems that this effect of the gas pressure or gas density cannot be explained by the theories known at that time.

However, at the fluidization conferences at Cambridge in 1978 it was already suggested by one of us (Mutters and Rietema, 1978) that gas adsorption on the surface of the solids might increase the cohesion forces between the solid particles and, hence, also that the elasticity modulus will increase with increasing gas pressure.

This does not seem unreasonable as both interparticle forces and gas adsorption have their origin in the van der Waals forces. At Eindhoven experimental evidence of this possibility was found with a specially designed tilting bed (Piepers *et al.*, 1984) which could be operated at increased gas pressure up to 10 bar. With this apparatus the yield curve of powders could be determined. The powder used was fresh cracking catalyst always at a porosity of 40% while argon was used as the fluidization gas.

In order to come as close as possible to the shear stress at zero normal stress a very small bed height of only 0.02 m was chosen, while, in order to reduce the normal stress σ_n at the bottom even further, an upward gas flow was applied at a velocity just below the incipient fluidization velocity. By varying this gas velocity the normal stress could be varied, while the corresponding shear stress could be measured by tilting the bed until shearing off occurs. The yield

Fig. 8. Variation of maximum homogeneous bed expansion with gas density for three different gases. Comparison with empirical correlation of Abrahamson and Geldart (1980a) (—). Gases: (■) H_2 , (Δ) N_2 , ($\circ$) Ar.

curves found are given in Fig. 9. From these curves it follows clearly that the cohesion constant (= shear stress at $\sigma_n = 0$) increases with gas pressure.

A theory on interparticle forces and on the effect of gas pressure on these forces was recently published by Cottaar and Rietema (1986). Their theory starts from the van der Waals forces between separate molecules as formulated by the Lennard-Jones potential. They could show that indeed—depending on the gas-solid system—the cohesion between particles could increase by a factor 2 or 3 for gas pressures up to 10 bar.

In dimensionless notation Cottaar and Rietema's theoretical cohesion is plotted in Fig. 10 vs the cohesion constant as determined by the tilting-bed method. A remarkably good correlation between the two can be observed.

CORRELATION WITH GAS DENSITY

In order to investigate the effect of gas pressure on expansion of the homogeneously gas-fluidized bed experiments were carried out with two powders, fresh cracking catalyst and polypropylene powder. The physical properties of the powders are given in Table 1. These powders were fluidized with different gases (see Table 2) and at pressures up to 15 bar.

The fluidization apparatus was a cylindrical glass column (i.d. = 0.082 m) about 1.5 m high. The fluid-

Fig. 9. Yield curves of fresh cracking catalyst in argon atmosphere at different gas pressures.

Fig. 10. Correlation of theoretical cohesion according to Cottaar and Rietema (1986) with empirical cohesion as measured in the tilting bed.

Table 1. Physical properties of the powders

Powder [†]	Skeletal density ρ_s (kg/m ³)	Internal porosity ϵ_i	Particle density ρ_p (kg/m ³)	Surface-volume mean diameter d_p (μ m)
FCC	2465	0.640	887	59.4
PP	918	0.389	561	76.0

[†] FCC = fresh cracking catalyst, PP = polypropylene.

Table 2. Physical properties of the gases

Gas	Density (kg/m ³) at 1 bar and 18°C	Viscosity (Ns/m ² × 10 ⁷) at 18°C
Hydrogen	0.089	88
Helium	0.1674	192
Methane	0.672	110
Neon	0.844	310
Nitrogen	1.174	173
Argon	1.674	220

ization gas was continuously recycled after passing through a dust filter. Two expansion vessels were part of the gas circuit to damp out small pressure fluctuations caused by the gas pump. When changing over from one gas to another the whole equipment was evacuated 3 times with subsequent refreshing of the gas phase.

From a graph of the bed height against the superficial gas velocity the bubble point velocity u_{mb} and the maximum stable bed height H_{mb} were determined (Fig. 11). Near the bubble point the fluidization velocity was carefully increased in small steps to avoid premature bubbling. The appearance of the first bubbles always resulted in small fluctuations of the bed height. H_{mb} was defined as the maximum bed height at which fluctuations did not yet occur. The bubble point porosity ϵ_{mb} at the maximum stable bed height H_{mb} was calculated from $\epsilon_{mb} = 1 - W/(H_{mb}A\rho_p g)$. The corresponding elasticity modulus E_{mb} was calculated from eq. (12).

It was assumed that the function E_{mb} can be conceived as the product of two independent functions, one:

$$f_1(\epsilon) = E_{mb} = E_{50} e^{-1.5(\epsilon_{mb} - 0.50)} \quad (16)$$

describing the dependency on porosity, and the other, $f_2(\rho_g)$, describing the dependency on the gas density.

The first function, $f_1(\epsilon)$, was derived experimentally at a pressure of 1 bar by applying different gases so that the critical bed expansion varied, assuming that at a pressure of 1 bar gas adsorption plays a negligible role (see Fig. 12). By applying eq. (16) the experimentally derived E_{mb} was converted into the corresponding elasticity modulus E_{50} at a porosity of 0.50 (see Tables 3 and 4). Thus, E_{50} probably is a measure for the forces acting at the contact points between the particles, while the porosity dependence is determined

Fig. 11. Example of H_{mb} and u_{mb} determination. Fig. 12. Plot of elasticity modulus E_{mb} vs bed porosity (fresh cracking catalyst) measured with six different gases at 1 bar. Gases: (+) CH₄, (Δ) N₂, (○) Ar, (■) H₂, (▲) He, (●) Ne.

by the number of contact points per unit volume of dispersed phase.

In Fig. 13 the calculated values of E_{50} are plotted vs the gas density for all six gases and all gas pressures. There appears to be a nearly linear relationship between E_{50} and ρ_g which is characteristic for the fresh cracking catalyst used in the experiments. It is determined by the physico-chemical interaction of the solid with the surrounding gas. The relationship shown in Fig. 13 seems to suggest that the amount of gas adsorbed increases with the gas density.

The results of the experiments carried out with polypropylene elaborated in the same way by means of eq. (16) are also indicated in Fig. 13. The characteristic curve of this solid is clearly different from that of fresh cracking catalyst.

It must be realized that these characteristic curves have been determined at room temperature. Since

Table 3. Results of experiments on homogeneous fluidization of fresh cracking catalyst with different gases

Pressure (bar)	Hydrogen						Helium					
	ρ_g (kg/m ³)	u_{mb} (cm/s)	H_{mb} (cm)	ϵ_{mb}	E_{mb} (N/m ²)	E_{50} (N/m ²)	ρ_g (kg/m ³)	u_{mb} (cm/s)	H_{mb} (cm)	ϵ_{mb}	E_{mb} (N/m ²)	E_{50} (N/m ²)
1	0.0843	0.80	71.2	0.511	0.850	1.00	0.1674	0.57	82.5	0.578	0.293	0.944
3	0.253	0.72	71.2	0.511	0.688	0.81	0.502	0.55	83.0	0.580	0.270	0.896
6	0.506	0.72	71.2	0.511	0.688	0.81	1.004	0.55	83.5	0.582	0.267	0.913
9	0.759	0.72	71.2	0.511	0.688	0.81	1.507	0.55	83.5	0.582	0.267	0.913
12	1.012	0.72	72.0	0.517	0.665	0.86	2.01	0.55	83.6	0.583	0.266	0.923
15	1.265	0.72	72.2	0.518	0.661	0.87	2.51	0.56	85.0	0.590	0.264	1.01
	Methane						Neon					
	ρ_g (kg/m ³)	u_{mb} (cm/s)	H_{mb} (cm)	ϵ_{mb}	E_{mb} (N/m ²)	E_{50} (N/m ²)	ρ_g (kg/m ³)	u_{mb} (cm/s)	H_{mb} (cm)	ϵ_{mb}	E_{mb} (N/m ²)	E_{50} (N/m ²)
1	0.672	0.70	75.5	0.538	0.560	0.99	0.844	0.50	91.0	0.617	0.182	1.05
3	2.015	0.73	77.0	0.547	0.573	1.13	2.532	0.50	93.0	0.625	0.173	1.13
6	4.03	0.83	79.3	0.560	0.686	1.69	5.064	0.55	96.0	0.637	0.196	1.53
9	6.05	0.91	81.0	0.570	0.782	2.23	7.596	0.58	97.5	0.642	0.213	1.79
12	8.06	0.94	82.4	0.577	0.802	2.55	10.13	0.63	99.8	0.651	0.240	2.31
15	10.08	0.98	83.9	0.584	0.838	2.95	12.66	0.70	104.0	0.664	0.276	3.23
	Nitrogen						Argon					
	ρ_g (kg/m ³)	u_{mb} (cm/s)	H_{mb} (cm)	ϵ_{mb}	E_{mb} (N/m ²)	E_{50} (N/m ²)	ρ_g (kg/m ³)	u_{mb} (cm/s)	H_{mb} (cm)	ϵ_{mb}	E_{mb} (N/m ²)	E_{50} (N/m ²)
1	1.174	0.632	83.7	0.583	0.351	1.22	1.674	0.590	87.0	0.600	0.278	1.25
3	3.522	0.665	85.4	0.591	0.371	1.45	5.022	0.650	90.4	0.615	0.310	1.74
6	7.044	0.790	87.5	0.601	0.495	2.25	10.04	0.720	94.5	0.631	0.349	2.50
9	10.57	0.850	91.2	0.618	0.522	3.06	15.07	0.850	98.0	0.644	0.453	3.93
12	14.09	0.875	92.7	0.624	0.535	3.44	20.01	0.940	101.0	0.655	0.522	5.34
15	17.61	0.950	95.5	0.635	0.594	4.50	25.10	1.03	104.0	0.665	0.593	7.04

Table 4. Results of experiments on homogeneous fluidization of polypropylene powder with different gases

Pressure (bar)	Nitrogen						Argon				
	u_{mb} (cm/s)	H_{mb} (cm)	ϵ_{mb}	E_{mb} (N/m ²)	E_{50} (N/m ²)		u_{mb} (cm/s)	H_{mb} (cm)	ϵ_{mb}	E_{mb} (N/m ²)	E_{50} (N/m ²)
1	0.58	68.9	0.512	0.278	0.33		0.56	69.5	0.515	0.257	0.32
3	0.62	71.7	0.530	0.289	0.45		0.70	73.0	0.538	0.352	0.62
6	0.72	73.0	0.538	0.367	0.65		0.79	75.5	0.554	0.408	0.92
9	0.78	76.0	0.557	0.386	0.91		0.88	78.0	0.568	0.468	1.30
12	0.82	77.7	0.566	0.411	1.11		0.96	79.0	0.574	0.538	1.63
15	0.84	78.5	0.571	0.419	1.22		1.00	81.0	0.584	0.552	1.95

physical adsorption of gas on to solid surfaces generally decreases at increasing temperature, we expect that the characteristic curves will also depend on temperature.

HETEROGENEOUS FLUIDIZATION

The foregoing theory concerns, of course, only the case of homogeneous fluidization. As nearly all applications of gas-solid fluidization, however, are in the region of heterogeneous fluidization one might wonder what the practical significance of the theory could be.

In heterogeneous fluidization the dense phase is frequently disturbed by passing bubbles, but will be stabilized again if the relaxation time τ_r of a distur-

bance is short enough, i.e. short compared to $1/f$ when f is the bubble frequency.

From the stability theory (Rietema and Mutsers, 1973; Mutsers and Rietema, 1977a) it follows that the rate of decline of a disturbance is given by e^{-at} , where

$$a = \frac{1}{\tau_r} = \frac{B}{2} \left(1 - \frac{V_c}{V} \right) \quad (17)$$

where V is the propagation velocity of the disturbance. As V might have any value between V_c and V_d a is > 0 .

$$B = \frac{150\mu}{\rho_p d_p^2} \left(\frac{1 - \epsilon}{\epsilon^3} \right). \quad (18)$$

Table 5. Results of experiments on heterogeneous fluidization of fresh cracking catalyst with different gases at a superficial gas velocity of 0.05 m/s

Pressure (bar)	δ (%)			ε_d			u_{bo} (cm/s)			f (s ⁻¹)		
	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar
	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar
1	6.9	7.1	7.2	0.480	0.489	0.524	4.53	4.72	4.70	3.06	3.9	4.3
3	8.1	7.1	7.4	0.489	0.510	0.544	4.50	4.67	4.61	3.70	4.5	6.1
6	8.1	7.8	8.5	0.495	0.531	0.579	4.54	4.60	4.45	4.13	5.7	8.9
9	8.3	8.7	8.8	0.504	0.548	0.600	4.50	4.50	4.35	4.69	7.3	11.4
12	8.0	9.4	10.4	0.513	0.568	0.618	4.44	4.38	4.22	5.02	10.0	12.9
15	7.4	10.3	10.0	0.518	0.578	0.629	4.47	4.31	3.96	5.42	10.2	14.0
	U_b (cm/s)			$\bar{D}_b$ (cm)			ε_{mb}			E_{50} (N/m ²)		
	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar
	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar	CH ₄	N ₂	Ar
1	66	66	65	2.22	1.83	1.65	0.538	0.583	0.600	0.99	1.22	1.25
3	56	66	62	1.82	1.55	1.13	0.547	0.591	0.615	1.13	1.45	1.74
6	56	59	52	1.65	1.20	0.75	0.560	0.601	0.631	1.69	2.25	2.50
9	54	52	49	1.44	0.93	0.58	0.570	0.618	0.644	2.23	3.06	3.93
12	56	47	41	1.33	0.65	0.50	0.577	0.624	0.655	2.55	3.44	5.34
15	60	42	40	1.24	0.63	0.43	0.584	0.635	0.665	2.95	4.50	7.04

As an example we consider the situation for the cracking catalyst-argon system at a pressure of 1 bar. For this system the bubble point porosity $\varepsilon_{mb} = 0.600$, the elasticity modulus E at this porosity is 0.278 N/m², and the solid density $\rho_p = 887$ kg/m³. From these data it can be calculated that $V_d = V_c = 0.0177$ m/s at $\varepsilon = 0.600$.

When fluidized heterogeneously at a superficial gas velocity of $u_o = 0.05$ m/s the dense-phase porosity ε_d proves to be 0.524 while the bubble frequency $f = 4.3$ s⁻¹ which indicates that the time between two bubble passages is 0.23 s.

From eqs (2) and (5) it follows that

$$V_c = \frac{\rho_p g d_p^2}{150\mu} \left[\frac{(3 - 2\varepsilon)\varepsilon^2}{1 - \varepsilon} \right]. \quad (19)$$

The porosity dependence of E is given by

$$E = 0.278e^{-1.5(\varepsilon - 0.60)}.$$

Hence, at $\varepsilon = 0.524$, these data give

$$V_c = 0.0105 \text{ m/s}$$

$$V_d = 0.0313 \text{ m/s}$$

$$B = 3488 \text{ s}^{-1}.$$

It follows that the relaxation time for a disturbance with a propagation velocity only a fraction larger than V_c —say $V = 0.0106$ m/s—is already < 0.06 s while the relaxation time decreases fast when the propagation velocity further increases. When, for example, $V \geq 0.0120$ m/s $\tau_r < 0.005$ s. Hence, it can be concluded that between two bubble passages the dense phase may be considered to be completely stabilized.

In the same apparatus as described before experiments were carried out with fresh cracking catalyst at higher superficial gas velocities so that most of the gas passed through the bed as bubbles.

Fig. 13. Correlation of E_{50} with gas density for six different gases and two solids (FCC = fresh cracking catalyst, PP = polypropylene). Gases: (+) CH₄, (Δ) N₂, (○) Ar, (■) H₂, (▲) He, (●) Ne.

By means of the collapse experiment (Rietema, 1967) we measured the dense-phase porosity ε_d , the bubble hold up δ and the superficial dense-phase velocity u_{do} . The bubble frequency f was measured by means of a capacity probe immersed in the bed at a height of 0.65 m above the gas distributor [see also Piepers *et al.* (1984)]. The superficial bubble velocity u_{bo} was calculated from the gas flow balance (Rietema and Oltrogge, 1968; Kunii and Levenspiel, 1969):

$$u_o = (1 + 2\delta)u_{do} + u_{bo}.$$

The average bubble diameter follows from^{*}

$$D_b = 1.5 \frac{u_{bo}}{f}. \quad (20)$$

The experiments were carried out at gas pressures up to 15 bar with three different gases: methane, nitrogen and argon, while the superficial gas velocity was

^{*} In the paper by Piepers *et al.* the bubble diameter was erroneously calculated a factor 4 too high.

Fig. 14. Correlation of D_b with μE_{50} at $u_0 = 0.05$ m/s and gas pressures up to 15 bar. Gases: (+) CH_4 , (Δ) N_2 , ($\circ$) Ar.

Fig. 15. Correlation of ϵ_d with μE_{50} at $u_0 = 0.05$ m/s and gas pressures up to 15 bar. Gases: (+) CH_4 , (Δ) N_2 , ($\circ$) Ar.

varied from 0.02 to 0.05 m/s. The results for a gas velocity of 0.05 m/s are given in Table 5.

From Table 5 it follows that the higher the E_{50} the smaller the bubbles. At higher E_{50} the dense-phase porosity increases and hence the cohesion of the dense phase decreases. This causes bubbles, which have become too large, to split sooner into smaller bubbles. That at higher dense-phase porosity bubbles remain of smaller size has also been noticed by other authors (Abrahamson and Geldart, 1980b; King and Harrison, 1980; Weimer and Quaderer, 1985).

In Fig. 14 the average bubble size and in Fig. 15 the dense-phase porosity have been plotted vs the product of E_{50} and the gas viscosity μ . These plots show remarkably good correlations.

The effect of gas viscosity seems rather surprising. There are, however, two possible explanations:

- Bubbles increase in size by means of gas extraction from the dense phase to the bubble phase as has been noticed by Rietema (1967) and Davies and Richardson (1966). This effect plays a role especially near the gas distributor but will decrease when the viscosity increases.
- Bubbles increase in size by coalescing with other bubbles. This coalescing is preceded and initiated by the coalescence of the bubble clouds

when the bubbles have approached each other close enough. At higher gas viscosity, however, the size of the clouds decreases and, hence, it can be expected that this coalescing of the clouds does not come into effect before the bubbles have approached each other more closely. This means that at a higher gas viscosity the rate of coalescing is smaller and hence the bubbles on average are smaller.

The two effects, one on the average bubble size and the one on the dense-phase porosity, both increase the mass transfer from the dense phase to the bubble phase and therefore seem to be of the greatest importance for the operation and design of fluidized-bed reactors.

CONCLUSIONS

The experiments prove that the elasticity quantified by the elasticity modulus E is a real and significant property of the powder bed which has its import both on homogeneous and heterogeneous fluidization. It is related directly to the interparticle forces.

The theory of Foscolo and Gibilaro (1984, 1987) is based on wrong premises and on a serious mathematical mistake as shown in Appendix II.

A final remark should be made: pure elastic deformations are always reversible and happen without energy dissipation. In hydrodynamics, however, flows and movements are always accompanied with energy dissipation. From these considerations it can simply be concluded that on the basis of hydrodynamics elastic forces do not exist.

NOTATION

a	damping factor ($1/\tau_r$), s^{-1}
A	cross-sectional area of fluidized bed, m^2
Ar	Archimedes number
A_{ss}	Hamaker constant, Nm
B	constant in eq. (13), s^{-1}
C_{ss}	constant in Lenard-Jones potential, Nm^7
d	diameter of particle, m
d_p	average particle diameter, m
D_b	average bubble diameter, m
E	elasticity modulus of powder structure, N/m^2
E_{mb}	value of E at the bubble point porosity, N/m^2
E_{50}	value of E at a porosity of 50%, N/m^2
f	bubble frequency, s^{-1}
F	slip force of dispersed phase on continuous phase, N/m^3
F_e	elastic force per unit surface area, N/m^2
F_i	interaction force, N
g	gravitational constant, m/s^2
g_c	apparent gravitational constant, m/s^2
h	height in fluidized bed, m
H	height of fluidized bed, m
H_{mb}	height of fluidized bed at bubble point, m
n	exponent in Richardson-Zaki equation
p	gas pressure, N/m^2

Re_t	Reynolds number for unhindered settling particles, $(\rho_g d_p u_t / \mu)$
r	intermolecular distance, m
r_{ss}	constant in Lennard-Jones potential, m
t	time, s
U_m	molecular energy, N m
u_t	terminal unhindered settling velocity of particle, m/s
u_o	superficial gas velocity, m/s
u'_o	approach gas velocity, m/s
u_{bo}	superficial bubble gas velocity, m/s
u_{do}	superficial dense-phase velocity, m/s
u_{mb}	minimum bubble point velocity, m/s
v_c	linear velocity of gas, m/s
v_d	linear velocity of solids, m/s
V_c	velocity of continuous waves, m/s
V_d	velocity of dynamic/elastic waves, m/s
V_{sa}	intermolecular potential, N m
W	total weight of fluidized bed, kg m/s ²
z	smallest distance between particles, m

Greek letters

α	tilting angle
α_c	critical tilting angle
δ	bubble hold up
ε	porosity
ε'	porosity perturbation
ε_d	porosity of dense phase
ε_i	internal porosity
ε_{mb}	porosity at bubble point
ζ	dimensionless distance between particles
ρ_g	density of gas, kg/m ³
ρ_p	density of solid particles, kg/m ³
ρ_s	skeletal density of particles, kg/m ³
μ	gas viscosity, N s/m ²
σ_n	normal stress in powder structure, N/m ²
τ	shear stress in powder structure, N/m ²
τ_c	powder cohesion, N/m ²
τ_r	relaxation time, s
ϕ	dimensionless interaction force

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APPENDIX 1: ROUGH ESTIMATE OF ELECTRICAL CONDUCTIVITY BY PARTICLE COLLISIONS IN A GAS-FLUIDIZED BED OF FREE FLOATING CONDUCTIVE PARTICLES

We consider a suspension of free floating conductive particles in an electrical field between two vertical electrodes. Each pair of two neighbouring particles is conceived as a capacitance. The capacity of a condenser existing of two parallel electrodes at a distance l is

$$C = \frac{A}{4\pi l} \frac{10^{-9}}{9} \text{ F}$$

where A is the surface area of the electrodes. In the case of the condenser existing of two neighbouring particles the effective area $A_p = \pi d_p^2/4$ and the effective distance is estimated to be $l = \epsilon d_p/4$.

Hence the capacity of a pair of particles becomes

$$C_p = \frac{d_p}{4\epsilon} \frac{10^{-9}}{9} \text{ F.}$$

We now consider a string of n particles in the direction of the lines of force, and hence in the direction perpendicular to the electrodes. The capacity of this string C_s , which arises through the cascade of particle capacities in the string, is given by

$$C_s = \frac{C_p}{n}.$$

If ΔV_o is the potential difference between the two electrodes, and L is the distance of these electrodes, then the potential difference ΔV_s over the string is

$$\Delta V_s = \Delta V_o \frac{n d_p}{L}.$$

By induction from the electrical field there arises on each particle of the string at one side a charge q_n and on the opposite side a charge $-q_n$, where

$$q_n = \frac{C_p}{n} \Delta V_s.$$

When in the string there occurs a collision between two neighbouring particles the number of capacities in the string decreases from n to $n-1$ and hence the charge increases from

$$q_n \text{ to } q_{n-1} = \frac{C_p}{n-1} \Delta V_s.$$

Hence an electric charge flows equal to

$$q_{n-1} - q_n = C_p \Delta V_s \left(\frac{1}{n-1} - \frac{1}{n} \right) \approx \frac{C_p \Delta V_s}{n^2}.$$

When there is a time Δt between two collisions there arises a small electric current i_s :

$$i_s = \frac{C_p \Delta V_s}{n^2 \Delta t}.$$

Suppose the number of collisions per particle and per second in the particle bed is m . Then the number of collisions per second in a string of n particles becomes nm , and the time Δt between two collisions in the string is $\Delta t = 1/nm$.

When per unit cross-sectional area there are N particles and hence N parallel particle strings the electric current density $i = Ni_s$ and the electric current through the electrodes becomes $I = Ni_s A$. With $m = u_o/d_p$ and $N = (1 - \epsilon)/d_p^2$ we obtain, after substitution of C_p and ΔV_s :

$$I = \frac{1 - \epsilon}{4\epsilon} \left(\frac{u_o}{d_p} \right) \frac{A}{L} \Delta V_o \frac{10^{-9}}{9} \text{ A.}$$

With $\epsilon = 0.5$, $A = 10^{-3} \text{ m}^2$, $u_o = 10^{-2} \text{ m/s}$, $L = 10^{-1} \text{ m}$, $d_p = 10^{-4} \text{ m}$ and $\Delta V_o = 20 \text{ V}$, we obtain $I \approx 0.5 \times 10^{-9} \text{ A}$.

This is a factor of 10^6 smaller than the measured electrical current through the particle bed (see Fig. 1).

APPENDIX 2: THE THEORY OF FOSCOLO AND GIBILARO

In a more recent paper Foscolo and Gibilaro (F + G) (1987) present a seemingly more exact derivation of the same result as in their paper of 1984. They express the net force, f (comprising gravity, buoyancy and drag), acting on a single particle as follows:

$$f = \frac{\pi d_p^3}{6} g (\rho_p - \rho_g) \left[\left(\frac{u'_o - v_d}{u_t} \right)^{\frac{4.8}{n}} \epsilon^{-3.8} - \epsilon \right]. \quad (\text{A1})$$

They now propose an extra force operating on what they call an upper layer of the packing. This force would be due to a porosity gradient around this layer. When f is the force on a single particle, this extra force on the whole layer then becomes

$$N_L \frac{\partial f}{\partial h} = N_L \left(\frac{\partial f}{\partial \epsilon} \right)_{\epsilon=\epsilon_o} \frac{\partial \epsilon}{\partial h} \quad (\text{A2})$$

where $N_L = [4(1 - \epsilon)]/\pi d_p^2$ is the number of particles in a layer. F + G suggest that this extra force is an elastic force which therefore resists any disturbance that might occur in the packing.

Let alone that N_L depends also on the porosity and hence should also belong under the differential operator it can easily be recognized that this extra force is not an elastic force but, on the contrary, will amplify any disturbance in the packing.

In the upper part of the disturbance shown in Fig. A1 $\partial \epsilon / \partial h$ is positive. $\partial f / \partial \epsilon$ of course is negative and hence the extra force is directed downward, which will cause a downward movement of the solids in this upper part of the disturbance. In the lower part just the opposite will happen and hence this disturbance will be intensified (see dotted line). On elaborating eq. (A2) from eq. (A1) ($f = 0$ for $\epsilon = \epsilon_o$) it is found that

$$N_L \left(\frac{\partial f}{\partial \epsilon} \right)_{\epsilon=\epsilon_o} \frac{\partial \epsilon}{\partial h} = - [3.2 g d_p (1 - \epsilon_o) (\rho_p - \rho_g)] \frac{\partial \epsilon}{\partial h} \quad (\text{A3})$$

F + G now put

$$\rho_p V_d^2 = + [3.2 g d_p (1 - \epsilon_o) (\rho_p - \rho_g)] \quad (\text{A4})$$

and add this force to the general interaction force F but erroneously without the minus sign.[†] Next they introduce these forces in the momentum balance of the solid phase.

After linearization of the continuity equation and the momentum balance of the dispersed phase they find that

$$\frac{\partial \epsilon'}{\partial t} - (1 - \epsilon_o) \frac{\partial v_d}{\partial h} = 0 \quad (\text{A5})$$

$$(1 - \epsilon_o) \frac{\partial v_d}{\partial t} + B [(1 - \epsilon_o) v_d + V_d \epsilon'] \frac{1}{V_d^2} \frac{\partial \epsilon'}{\partial h} = 0. \quad (\text{A6})$$

[†] Apart from the criticism of the original paper of (F + G, 1984) on this subject that we have already formulated in paragraph 3, F + G also make a serious error in this paper. In their eqs (25) and (27) for the additional force they also lose a minus sign. Equation (25) should read

$$\partial f = - \frac{A}{N_s} \left(\frac{\partial p'_{h,i}}{\partial h} \right) \Delta h$$

where $p'_{h,i}$ is the disturbance in the "equilibrium" gas pressure resulting from the bed compression.

Fig. A1. Example of a porosity disturbance in a fluidized bed.

By differentiating eq. (A5) with respect to t and eq. (A6) with respect to h and combining these equations to eliminate v_d , they finally obtain the single equation for voidage perturbation in the bed, which is the same as ours and that of Wallis (1969):

$$\frac{\partial^2 e'}{\partial t^2} \downarrow V_d^2 \frac{\partial^2 e'}{\partial h^2} + B \left(\frac{\partial e'}{\partial t} + V_c \frac{\partial e'}{\partial h} \right) = 0 \quad (\text{A7})$$

From eq. (A7) it follows that there will be stability at all porosities for which $V_d > V_c$.

As said already $F + G$ have lost the minus sign in eq. (A3). This would mean that the sign should be reversed at all places indicated by an arrow ($\downarrow$) in eqs (A4), (A6) and (A7). In the correct eq. (A7) the minus sign would have become a plus sign with the consequence that there will be instability at all porosities above the packed-bed porosity!