

## Fluid Beds: At Last, Challenging Two Entrenched Practices

Arthur M. Squires, Mooson Kwauk, Amos A. Avidan

In 1926, a fluid bed gasifying lignite sent fuel gas to Otto gas engines that compressed ammonia synthesis gas for I. G. Farben's Leunawerke, near Leipzig, Germany. On 25 May 1942, at Standard

or burning wastes, drying solids, cracking hydrocarbons, and converting gases catalytically (Table 1).

These two practices stand at the threshold of opportunities that few could

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**Summary.** Originating in the 1920's and 1930's, two distinct fluidization arts have emerged, one for treating coarse solids and the other for fine powders. Fluidization research has tended to focus on bubbling beds of coarse solids, but designers of such beds for burning coal have learned to appreciate the importance of combustion of fine char particles in the freeboard. Designers of successful processes for powders have focused on bubble suppression. Since about 1980, combustion fluid beds of both types are challenging the conventional pulverized-coal boiler; they provide better means for controlling emissions from the combustion of high-sulfur fuels. Progress in the "bubbleless" fluidization of fine powders is increasing the fluid bed's competitiveness with the fixed-bed catalytic reactor. Efforts to advance the fluid bed for catalysis, besides increasing gas velocities beyond levels that most researchers have used in the past, must include systematic study of the level of fine particles smaller than 40 micrometers.

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Oil of New Jersey's Baton Rouge refinery, a fluid bed began producing ingredients for 100-octane aviation gasoline: 87-octane "base stock" for blending with iso-octane and tetraethyl lead, and isobutylene for reaction with isobutane to yield iso-octane. Thus were born two distinct fluidization practices: a "German" practice for treating coarse solids, and an "American" practice for dealing with fine powders. Both have recorded successes, such as roasting ores, calcining carbonates and hydroxides, calcining

have imagined as recently as 1980. They are just now penetrating the market for large electricity-station boilers, and our understanding of the fine-powder practice for catalytic conversion of gases is approaching a maturity that may often defeat the fixed bed in future developments.

Some 50 companies in 25 countries now offer atmospheric-pressure fluid bed combustion (AFBC) of coal (1). In the People's Republic of China, more than 2000 AFBC boilers or other devices serve agricultural communes, industry, and electricity stations (2). In the West, nearly 300 units supply heat to food and paper industries, to municipalities in Scandinavia for district heat, to oil companies using steam for secondary oil recovery, and to farms in England for drying grass (3, 4). Operating experience

is accumulating that underscores AFBC's advantages. It can burn low-grade fuels, such as oil shales and coals high in mineral matter, that are unsuitable for combustion in the conventional pulverized-coal (PC) boiler. Indeed, some AFBC designs can switch fuels over a wide range of quality, burning municipal waste, peat, or wood as readily as coal (5). AFBC can use limestone to capture sulfur during the combustion process, and designs that afford two-stage combustion (6) can greatly reduce emissions of nitrogen oxides. The U.S. electricity industry, through its Electric Power Research Institute (EPRI), has closely followed AFBC developments (7). Figure 1 illustrates two AFBC boilers—one employing coarse solids and the other, fine powders—that will soon generate electricity. Both are retrofits for old stations, and their owners will enjoy an increase in electricity output along with an absolute reduction in emissions of sulfur dioxide and a relative reduction in nitrogen oxides (3). There are operating advantages as well. For example, PC boilers sometimes explode, whereas an AFBC probably cannot (at least, it is hard to imagine how). An AFBC can be banked overnight and restarted within minutes, its bed material having lost little in temperature. It can change load quickly (1). Heat flux to steam tubes (8) is comparable in present-day AFBC and PC designs (9). However, the radiant flux reaching tubes from PC combustion varies, and designers must consider the possibility that at some point the flux could attain an extreme level that would burn out tubes. Finally, large AFBC units (10) are costing less to build than traditional PC designs.

### Early Fluidization Arts

Fritz Winkler invented the first successful fluid bed for coarse solids (11). In December 1921, he saw how to design a chemical reactor employing a bed of particles levitated and set in boiling motion by a rising current of gas. If a gas is introduced into the bottom of a container of a coarse powder and the upward gas velocity is gradually increased, after a time the particles will lift and begin to

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Arthur M. Squires is University Distinguished Professor, Department of Chemical Engineering, Virginia Polytechnic Institute and State University, Blacksburg 24061. Mooson Kwauk, one of the approximately 370 members of the Chinese Academy of Sciences, is professor and director of the Academy's Institute of Chemical Metallurgy, Beijing, People's Republic of China. Amos A. Avidan is a senior research engineer, Mobil Research and Development Corporation, Paulsboro, New Jersey 08066.

Table 1. Major applications of heterogeneous catalysis in fluid beds.

Process	Licensors	Status
Fluid catalytic cracking (1942)	Exxon, UOP, Kellogg, Shell, Texaco, others	$10 \times 10^6$ barrels per day, >350 units
Phthalic anhydride (1945)	Sherwin-Williams, Badger, others	$0.3 \times 10^9$ pounds per year (United States)
Fischer-Tropsch synthesis (1955)	Kellogg, Badger	3 units (Sasol I), 16 units (Sasol II and III)
Chlorinated hydrocarbons and chlorine (early 1950's)	Badger, Shell, Uhde, PPG, others	Large number of units
Acrylonitrile (1960)	Sohio-Badger	$>6 \times 10^9$ pounds per year, >50 units
Polyethylene (high density, 1968) (low density, 1977)	Union Carbide	>15 units

boil. At a certain gas flow, an upward drag force just cancels the downward pull of gravity; particles become free to move relative to one another. A further increment of gas forms bubbles that rise at high speed through a fluidized particle mass (12).

Figure 2 illustrates generally how the volume fraction of a coarse solid varies with fluidizing gas velocity. Schematic equipment diagrams illustrate applications of the coarse solid fluidization practice. In the early 1960's, the British National Coal Board (following Douglas E. Elliott's lead) and Maoming Petroleum Company (Guangdong Province, China) almost simultaneously began AFBC studies using coarse solids. The latter's interest was utilization of oil shale fines.

Warren K. Lewis and Edwin R. Gilliland of Massachusetts Institute of Tech-

nology were the first to fluidize fine powders (13). In December 1938 and throughout 1939, they blew fluid catalytic cracking (FCC) powder upward through pipes at gas velocities from minimum buoyancy to about  $3 \text{ m sec}^{-1}$ . In mid-1940, a consortium led by Standard Oil of New Jersey and the M. W. Kellogg Company used Lewis and Gilliland's data to design a fluid-bed substitute for a pilot cracking reactor of another type, whose performance was disappointing (14).

Figure 3 illustrates the variation of volume fraction with gas velocity for a fine powder, such as the catalyst used in FCC. Two differences between the curves of Figs. 2 and 3 are important to the engineer. (i) In Fig. 3, bubbles do not appear after a gradually increasing flow of gas just buoys up the powder; rather, the powder expands uniformly over an

interval between a minimum buoyancy and a minimum bubbling velocity. The powder dilates before bubbles appear. (ii) In Fig. 2, the decline in solid volume fraction is more precipitous for a proportionate increase in gas velocity.

FCC powder is one of the most significant and fascinating artifacts of engineering technology. It is difficult to suggest a common material that bears some physical resemblance to FCC powder. Sugar and salt are too coarse. Flour is too fine and too cohesive. FCC powder is somewhat like cement powder but much more freely flowing. If a tightly lidded glass jar about half full of FCC powder is rotated rapidly, the powder assumes a liquid-like character. Rotation shears the powder, causing it to dilate; the powder therefore "imbibes" gas, creating thin films of gas that separate the particles—a gas-solid emulsion. If the jar is then held upright and shaken gently, the powder's surface sloshes back and forth for a number of seconds, during which gravity slowly pumps gas from the powder.

Bagnold (15) measured the dispersive pressure of an array of particles under shear and showed that it declines slowly with increase in particle-to-particle distance in a liquid but declines sharply to a negligible value when particles in a gas have moved slightly apart. We use "Bagnold distance" to denote the particle separation at the limit of shear's influence. Any array of particles, except for a special construction (such as a symmetrical pile of cannon balls), will display

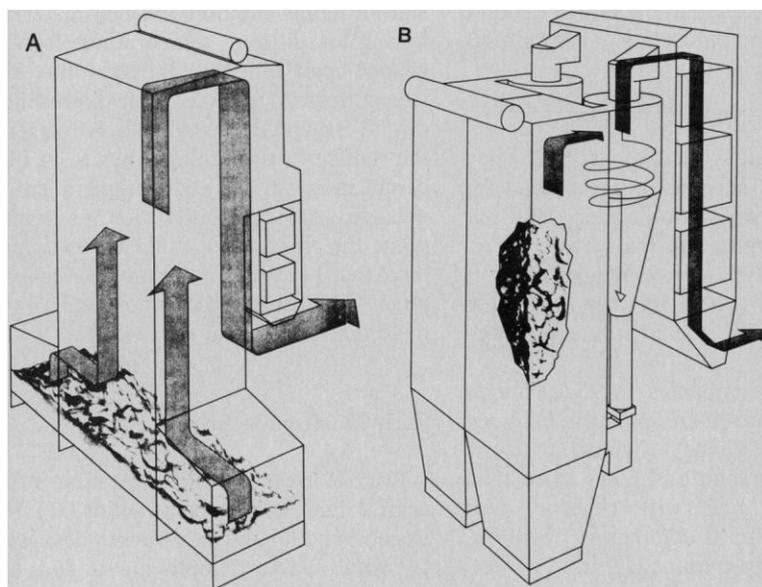


Fig. 1. (A) Atmospheric fluid bed combustion (AFBC) boiler in the German tradition for Northern States Power Company's Black Dog station, 15 miles south of Minneapolis, rated at 100 MW (electric) (3). Its fluid bed is about 1 m deep and is traversed by horizontal steam tubes to remove combustion heat. Bed material is coarse—a sand or a gravel—primarily composed of particles derived from limestone ( $\text{CaO}/\text{CaSO}_4$  in a ratio between about 0.5 and 1.0); conversion of  $\text{CaO}$  by reaction with  $\text{SO}_2$  and  $\text{O}_2$  is less than 50 percent because  $\text{CaSO}_4$  forms an impervious skin on a core of unreacted  $\text{CaO}$ . Superficial gas velocity (calculated at the temperature and pressure of the bed as if it were empty of both solid and tubes) is about  $3 \text{ m sec}^{-1}$ . Gas leaving the bed carries dust (fly ash, coal char, and the finer  $\text{CaO}/\text{CaSO}_4$  particles); equipment not shown in the drawing captures dust and recycles some of it to the bed, in an amount roughly twice that of coal feed. The recycle affords better opportunity for utilizing fine particles of lime and char created by processes of fragmentation and attrition. A large fraction of the fixed carbon in coal burns in the freeboard (87). (B) An AFBC boiler in the American tradition for Colorado-Ute Electric Association, Incorporated's Nucla station, rated at 110 MW (electric) (3). The circulating fluid bed (CFB) is a fine  $\text{CaO}/\text{CaSO}_4$  powder that is blown continuously into two cyclone dust separators; standpipes beneath the cyclones return powder to the bottom. Details of the design of Nucla's heat-exchange surfaces

are not available, but it is doubtful that any steam tubes are fully surrounded by fluidized solids. Membrane walls, formed by butt-welded tubes, enclose the bed. The heat-transfer surface may include butt-welded, vertical panels that extend inwardly from the walls but do not subdivide the bed into compartments. Recirculation of powder is 200 times coal feed; without such recirculation, no bed would form at Nucla's superficial gas velocity ( $6 \text{ m sec}^{-1}$ ). The fine limestone particles afford better reactivity than the larger particles at Black Dog; Nucla expects 60 percent conversion of  $\text{CaO}$  to  $\text{CaSO}_4$  at a slightly better sulfur retention than Black Dog's. [Reprinted from *EPRI Journal*]

variations in particle concentration. A region of low concentration becomes a short circuit for an upward flow of gas, and when the gas has diluted FCC powder a little beyond the Bagnold distance (16) the region consolidates itself into a nearly perfect Taylor bubble, with its characteristic mushroom-cap shape (17). The bubble's dynamics are similar to those of a bubble in an inviscid liquid; its velocity of rise, in general, is far beyond the gas superficial velocity at minimum bubbling. The bubble subjects nearby particles to intense shear and maintains their fluidity by keeping them separated at the Bagnold distance.

A column of fluidized powder exerts the same hydrostatic head as a column of liquid of the same specific gravity. With aeration to prevent collapse of interstitial

gas films as gas pressure increases, a standpipe can deliver solid at a pressure elevated by the head developed by the column of powder.

The first FCC units (I in Fig. 3) used an upflow design: catalyst powder and oil vapor entered at the bottom of a fluid bed, and catalyst and cracked products emerged together at the top. But FCC's developers soon discovered a "meniscus effect." Sometimes upward flowing catalyst would form two distinct regions: a surging interface, resembling the surface of a boiling liquid, separated a dense region in the lower part of a vessel from a dilute region above. Whether a meniscus is present depends on the rate of catalyst flow. If the flow is large and the only exit for powder is at the top, only a dense phase is present. If catalyst flow is small,

or if a standpipe is provided to withdraw powder downward from the dense phase, a dilute phase appears. Catalyst entering the bed must at least replace the carryover; to reduce losses, designers usually provide cyclone dust separators that return powder to the bed. Most designers specify a distance between the meniscus and the cyclone inlet—the freeboard—that is at least the "transport disengaging height" (18), so that splash from the surging meniscus does not contribute to carryover. In such a design, the flow of catalyst into the cyclones is the saturation upward conveying capacity of the gas. After appreciating the meniscus effect, inventors conceived the downflow design (II in Fig. 3), which is a configuration resembling Winkler's fluid bed.

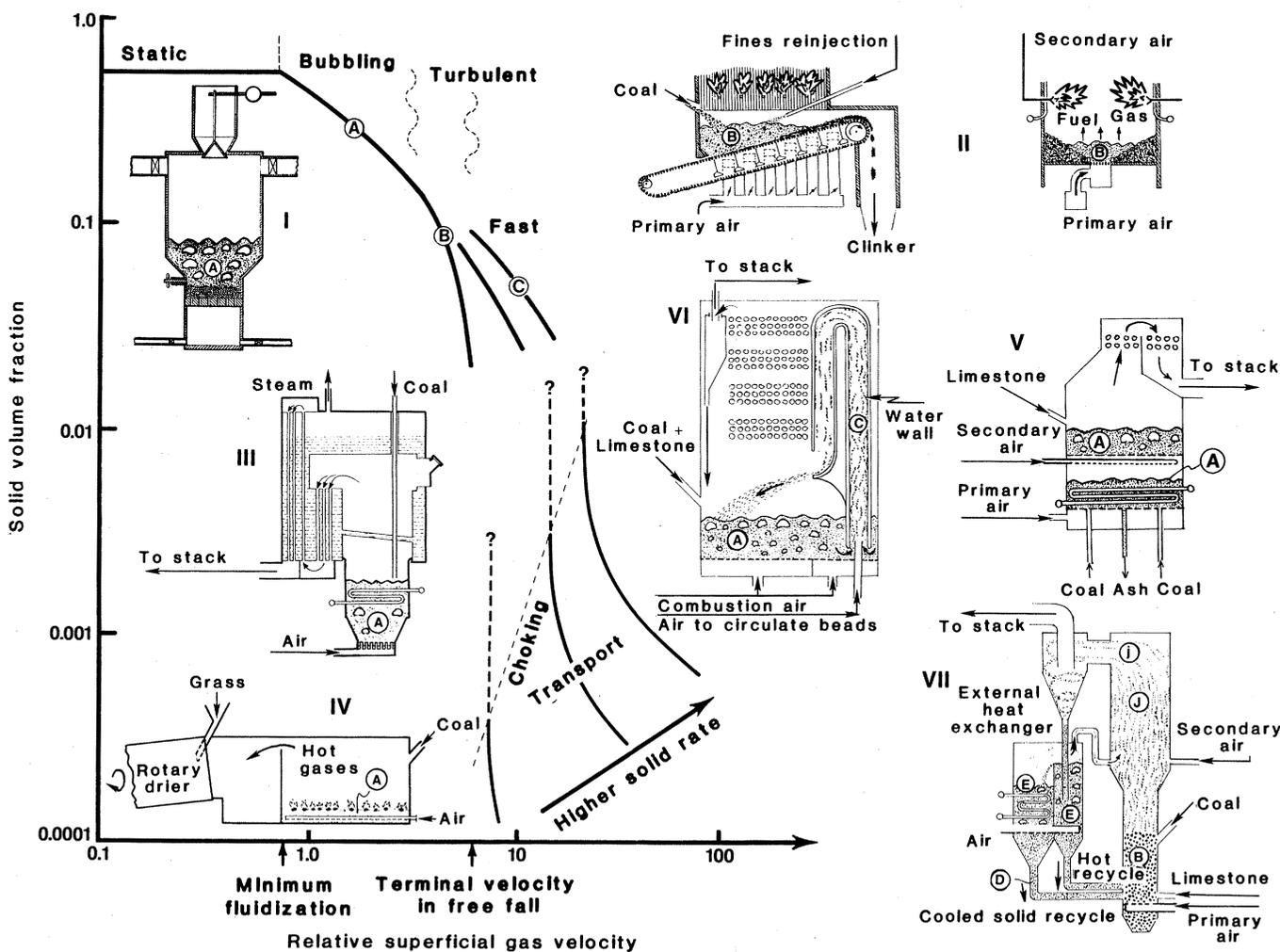


Fig. 2. Illustrative fluidization regime diagram for a coarse solid; solid volume fractions are averaged over the cross section of the column. Staub (34) observed the transition from bubbling to turbulence; Stromberg (60) reported data for dense fluidization beyond the terminal velocity. Matsen (88) gives an expression for transport; a vertical dotted line indicates choking (61). (I) Adapted from Winkler's first patent (11). (II) Ignifluid boiler, which uses a gasification bed to produce a fuel gas for secondary combustion in conventional boiler space. The bed operates at about 10 to 20 m sec<sup>-1</sup> and 1200° to 1400°C; under these conditions, ash matter forms walnut-sized agglomerates that remain freely fluidized until, by eddy diffusion, they join a sticky pad of clinkers that forms on a traveling grate (42, 85). (III) Vertical shell boiler (89), an early prototype for the Black Dog design and many offerings of AFBC boilers in small sizes. (IV) Burning lumps of washed, sized coal floating on a shallow fluid bed of sand (89, 90) supply hot gas for drying grass. (V) Multibed design (91), with a lower bed of inert solid operating at a higher temperature than an upper bed of CaO/CaSO<sub>4</sub>. (VI) CFB design circulating tough beads of alumina and employing fine limestone (92). Recirculated fines amount to about 10 times coal feed; bead recirculation is about 100 times coal feed. (VII) Hybrid with a stationary, turbulent combustion zone of river sand; a circulating fine powder delivers heat to an external, bubbling fluid bed boiler (3, 93). See Fig. 3 for significance of letters beyond C.

Progress in Fluidization Arts

Operators of fluid crackers soon became aware that the dense phase in a bubbling bed is a region of gas backmixing (19): bubbles drag solids upward, and a backflow of solid elsewhere carries interstitial gas downward. This is often highly converted gas (essentially at thermodynamic equilibrium) because the gas creeps upward in laminar flow, in superb contact with the powder. In the catalyst regenerator of an FCC unit, air burns carbon from the catalyst. An early dis-

covery (20) was that a high proportion of samples of gas withdrawn from a bubbling regenerator showed very little oxygen; these samples came from the dense phase. A few samples showed oxygen contents higher than those in stack gas; these came from bubbles. Operators discovered that performance of FCC units improved when they drove their units to higher throughputs. They learned that fluidization is better at higher fluidizing gas velocity and higher level of the finer sizes of catalyst (21). FCC powder has a median diameter of about 55  $\mu\text{m}$ , with a

range of diameter between about 10 and 120  $\mu\text{m}$ . If the fraction below 40  $\mu\text{m}$  is too small, bubbles grow to large sizes, burst at the bed surface, and eject solids, which increase carryover. Restoring fines to the bed paradoxically reduces carryover, probably through both an improvement in catalyst fluidity (22) and an increase in the time for loss of fluidity when gas flow is cut off (23).

Lewis and Gilliland called fluidization with a meniscus "batch fluidization" and extended their work on "continuous" fluidization well into the fast re-

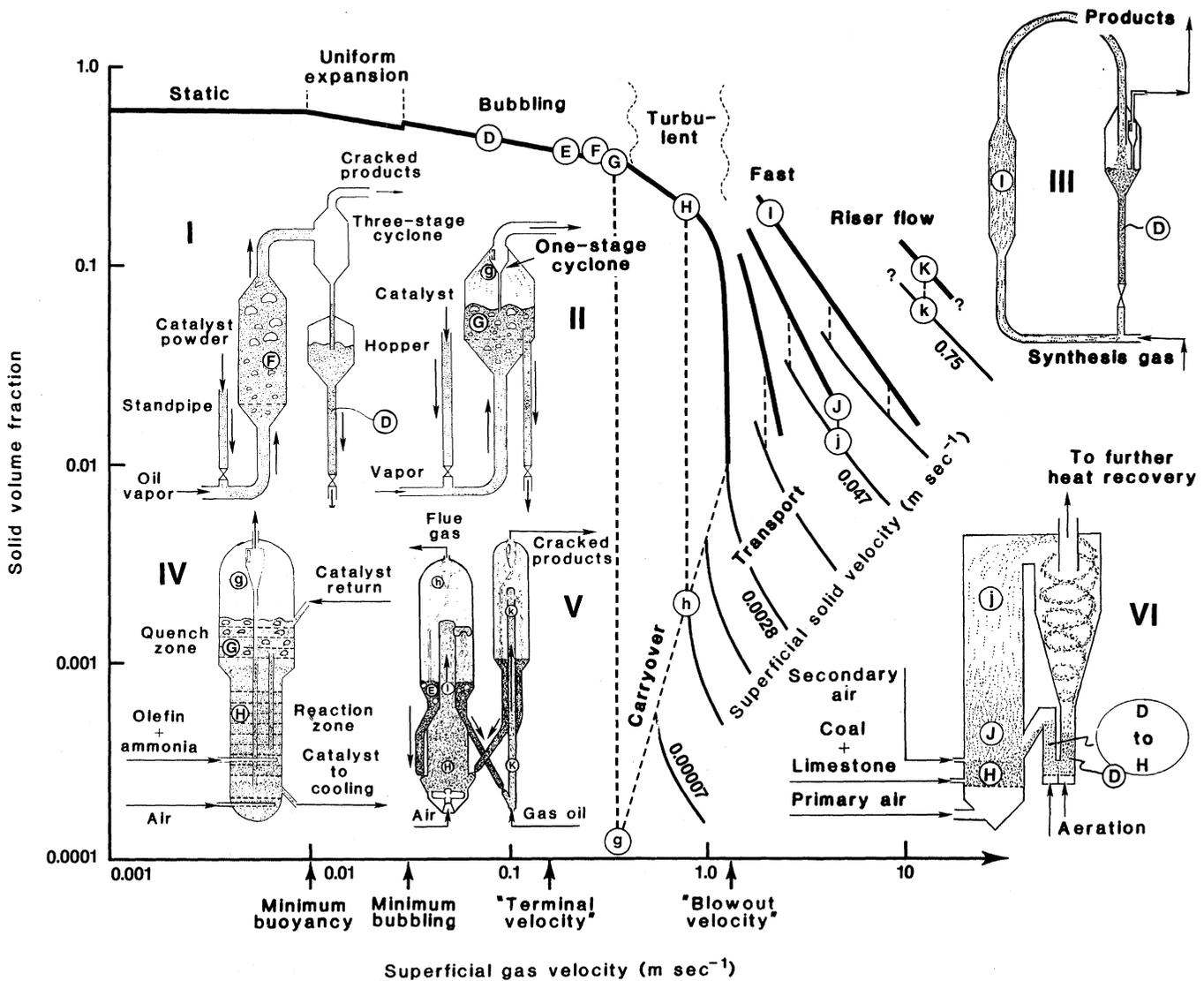


Fig. 3. Illustrative regime diagram for a fine powder; solid fraction is a column cross-sectional average. Vertical dotted lines connect two regimes that can coexist in a column; see Fig. 5A for typical vertical profiles of solid volume fraction. For a given solid rate, a broken line indicates interdicted solid fractions: an experimenter cannot create an extended vertical region at a solid fraction on the broken line. Figure 5, B, C, and D, illustrates how solid fraction can vary horizontally across a column; cross-sectional averages are specific for a given column geometry and may depend on the content of fines smaller than 40  $\mu\text{m}$ . The available data for velocities immediately beyond the blowout velocity are from cold CFB models 102 and 152 mm in diameter (35, 40, 47, 94), which is too small to give us confidence in our picture of how transport curves collide with curves for the fast regime. Data from larger models will soon become available (50). Relatively little is known about the riser flow regime (95, 96). (I) Upflow design of original fluid catalytic cracker (FCC model I). (II) Downflow design with a meniscus (model II). The first downflow FCC's (in the 1940's) operated at about 0.4  $\text{m sec}^{-1}$ , with a dust loading of about 1.7  $\text{kg per cubic meter}$  of gas entering a single-stage cyclone. Before 1955, FCC regenerators, similar to II in mechanical arrangement, operated at about 0.75  $\text{m sec}^{-1}$  (H) with about 70  $\text{kg m}^{-3}$  (h) dust loading into a three-stage cyclone (21). (III) CFB for Fischer-Tropsch synthesis at Sasol; steam tubes within the bed remove reaction heat. (IV) Staged turbulent bed for acrylonitrile; staging is advantageous if the fines content of catalyst is small. Some acrylonitrile is made in turbulent beds without staging (43). (V) Modern fluid cracker (Mobil-UOP) with a riser cracking zone and a CFB catalyst regenerator. (VI) CFB boiler; walls comprise butt-welded steam tubes.

gime of Fig. 3 (24), with high rates of solid feed to the bottom of a column. They described a "blowout velocity," that is, "a gas velocity sufficient to blow all or substantially all of the solid material out of the reactor in a relatively short time, provided no fresh solid material be introduced during this time" (24). Beyond the blowout velocity, solid flow rate has a major effect on the solid fraction in the dense phase.

In the early 1950's, the M. W. Kellogg Company used the fast regime in the design of circulating fluid bed (CFB) reactors for South Africa's Fischer-Tropsch synthesis plant at Sasolburg (25). These reactors afford high conversions of hydrogen and carbon monoxide to liquid hydrocarbons that can be refined to yield gasoline. The conversions attest to excellent contact between synthesis gas and catalyst, which is all the more striking in view of the failure of bubbling fluid bed reactors for this synthesis at Brownsville, Texas (13). In the late 1960's, Lothar Reh and colleagues at Lurgi Gesellschaft developed a CFB calciner for the aluminum industry (26) that has evolved into the CFB boiler designs that entered commerce in about 1980 (27).

### Fluidization Dynamics

Geldart (28) classifies granular solids into groups (Fig. 4). A fine powder (in group A) is "aeratable" it imbibes gas to form an emulsion phase characterized by a long defluidization time. As Fig. 3 illustrates, blowout velocity is far greater than terminal velocity in free fall. The powder forms a fluid bed having great retentivity for fines that are far smaller than the average particle size. Fluidization does not separate a powder according to size, and carryover reflects the full distribution of sizes in the powder (29).

In contrast, a sandy solid (group B) defluidizes instantaneously if gas flow is stopped; minimum buoyancy and minimum bubbling velocities are the same. Blowout and terminal velocities also coincide. The ratio of highest to lowest operable gas velocity is smaller for group B than for group A solids. Fluidization classifies the solid by size: carryover does not reflect the full range of sizes of particles in the bed but is enriched in particles with terminal velocities below the fluidizing-gas velocity. Over time, such particles are stripped from the bed; for many of the AFBC devices illustrated in Fig. 2, the dwell time of a fine particle of carbon in a bed of group B solid is an

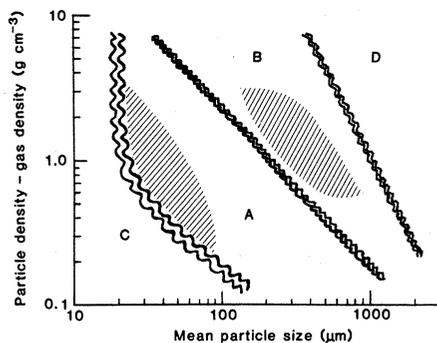


Fig. 4. Geldart's classification of granular solids (28). Shaded regions roughly characterize the group A aeratable powders and the group B sands that appear in most of those commercial applications in which designers can exercise control over particle size. Group C is too cohesive for bubbling fluidization; group D is spoutable. The boundary between A and B solids shifts to the right with increasing pressure.

important parameter affecting combustion efficiency (30). Like group A powders, group B solids fluidize better if present in a wide range of particle sizes (31): there is less tendency, for example, for a defluidized shoulder to form at the bottom of a bed next to the wall.

Beds of solids particulate fluidized by a rising current of liquid (32) generally exhibit smooth expansion. Fluid volume fraction,  $\epsilon$ , is conveniently represented by the empirical Richardson-Zaki expression (33):

$$U/U_T = \epsilon^n$$

where  $U$  is fluid velocity and  $U_T$  is the calculated terminal velocity of a particle in free fall. The exponent  $n$  varies from 3.65 at low fluid Reynolds number (based on particle diameter) to 4.65 at

high Reynolds number. The Richardson-Zaki expression represents fairly well the expansion of a bed of group B solid aggregatively fluidized by a gas (34), although the exponent  $n$  varies with gas velocity, tending to values beyond 3 in a bubbling bed of 650- $\mu\text{m}$  particles and below 2 in a turbulent bed. The Richardson-Zaki expression also describes the expansion of a bed of a gas-fluidized group A powder, if the powder's calculated terminal velocity is replaced by a far higher, empirically determined cluster terminal velocity,  $U_T^*$  (35, 36), which one might formally view as a terminal velocity that reflects the aggregation or clustering of the powder. In one series of experiments, typical fine powders exhibited  $U_T^*$  values between 12.2 and 38.7  $\text{m sec}^{-1}$  in the bubbling regime and between 2.8 and 6.3  $\text{m sec}^{-1}$  in the turbulent regime. The exponent  $n$  varied between about 8 and 10 in the bubbling state and between about 4 and 6 in turbulence. These trends in  $U_T^*$  and  $n$ , like similar trends in gas-liquid systems (37), reflect the finer scale in the demixing of gas and powder in the turbulent regime. In contrast with the bubbling regime's easily identifiable, often large bubbles, turbulence is characterized by "a state of continuous coalescence [of bubbles]—virtually a channeling state with tongues of fluid darting in zig-zag fashion through the bed" (38). The visual appearance of a column of fine powder does not change with the transition from turbulence to the fast regime, although both  $U_T^*$  and  $n$  now become functions of solid recirculation rate (35).

The transition from the bubbling to the turbulent fluidization regime is analo-

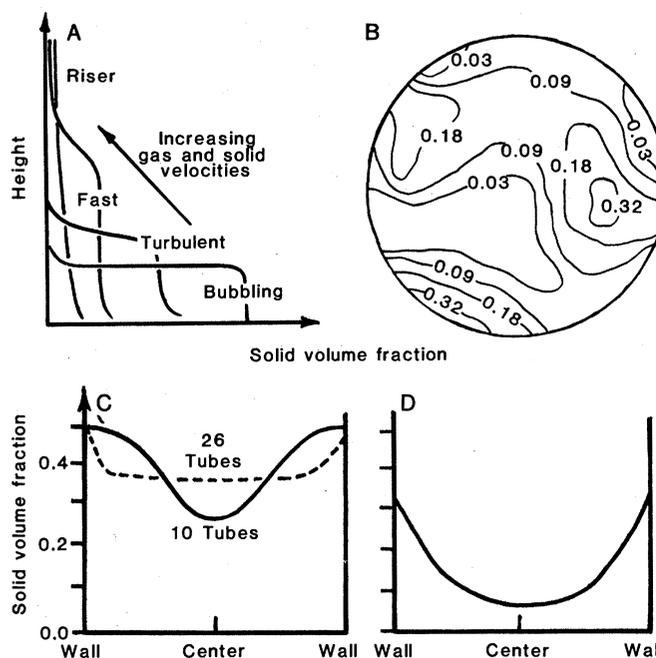


Fig. 5. Characteristic profiles of solid volume fraction in beds of group A powders. (A) Vertical profiles. (B) Contours of constant solid volume fraction in an FCC riser cracking zone (50); the curves illustrate demixing at a gross scale. (C) Horizontal profiles inferred from inductance probe exploration of a bubbling bed in a 747-mm vessel fitted with 47.6-mm vertical tubes (97). (D) Profile inferred from x-ray observations of the fast regime in a 152-mm column (98); the demixing of gas and solid occurs at a relatively fine scale.

gous to the well-known transition from the bubbly regime to the froth or churn-turbulent regime in upward flow of gas and liquid (39). In a column of small diameter, the transition can easily be recognized by following the fluctuations in a manometer that gives the pressure gradient over a portion of the column (34, 40, 41); in larger equipment, the transition can be inferred from the change in the exponent  $n$  in the Richardson-Zaki expression (35, 43, 44). For FCC catalyst—the prototypical group A powder—the transition is sharp (40); for some powders, however, the transition may occur over a range of velocities.

In upward pneumatic transport, particles also tend to cluster. The phenomenon can be readily seen in high-speed motion pictures (45) and deserves more experimental study than it has yet received (46). Formally, we may view the denser fluidization regimes as comprising systems in which transient clusters of relatively large, dense aggregates of particles are dispersed in a dilute continuum of sparsely distributed, smaller clusters. The denser clusters tend to move upward by a diffusive mechanism from a relatively dense region in the lower section of a column. When they arrive at a higher region, where the average solid fraction is lower, gravity pulls them downward toward the lower region. Dynamic equilibrium calls for equality of the fluxes driven by diffusion and gravity (47):

$$\xi \frac{d}{dz} \rho_s f \varphi_a = \omega \Delta \rho (\varphi_a - \varphi) f \varphi_a$$

where  $\xi$  and  $\omega$  are proportionality constants descriptive of the respective influences of diffusion and gravity on cluster motion;  $f$  is the volume fraction of clusters;  $\rho_s$  is the true density of particles;  $\Delta \rho$  is the density difference between particles and gas; and  $\varphi_a$ ,  $\varphi^*$ , and  $\varphi$  are solid volume fractions: respectively, the limiting fraction in the dense phase, the limiting fraction in the dilute phase, and the average value at distance  $z$  measured in the vertical direction. Noting that

$$\varphi = f \varphi_a + (1 - f) \varphi^*$$

one finds

$$\frac{\varphi_a - \varphi}{\varphi - \varphi^*} = \exp [-(z - z_0)/z_0]$$

where  $z_0$  is a characteristic length:

$$z_0 = \frac{\xi \rho_s}{\omega \Delta \rho (\varphi_a - \varphi^*)}$$

Plots of solid fraction versus distance give S-shaped curves (Fig. 5A) that accord with experience (47, 48). At the top of a column, solid fraction tends to  $\varphi^*$ ; at

the bottom, to  $\varphi_a$ . For small values of  $z_0$ , the inflection is abrupt, representing a distinct meniscus between dense and dilute regions. For large  $z_0$ , the meniscus is diffuse. Li and his co-workers (49) have given correlations for  $\varphi_a$  and  $\varphi^*$  based on data from a 102-mm column, but the correlations must be viewed with caution until data from larger columns become available (50). The vertical location of the meniscus in a column, whether in bubbling, turbulent, or fast fluidization, depends simply on the pressure difference imposed across the column (51); in all of the fluidization regimes, the pressure gradient in a column simply reflects the hydrostatic head of the solids (52).

As Fig. 5, B, C, and D, illustrate, the column cross-sectional average solid volume fractions of Figs. 2 and 3 may reflect, in some instances, extreme variations in volume fraction at various points across a given cross section. Data of this type are rare (43, 53). In the bubbling regime, especially when fines content is not adequate, introduction of vertical surface makes the solid fraction more uniform (Fig. 5C), lowers the cross-sectional average (54), increases gas residence time, and improves gas-solid contacting (55). Juxtaposition of Fig. 5, C and D, suggests that the variation in volume fraction in the fast regime may also be controlled, should this be desirable, by introducing vertical surface. Extreme variations in solid volume fraction across a fast-fluidized column have been reported (56), and we speculate that adding fines may be another approach to reducing such variation. Operators of fluid catalytic crackers know that adding fines reduces pressure fluctuations in a riser cracking zone (57), perhaps providing better dispersion of powder than is displayed in Fig. 5B. We do not know how and where on Fig. 3 the fine scale demixing in the fast regime merges into the gross demixing of the riser. Perhaps there is a transition, analogous to the transition in gas-liquid systems from churn-turbulent to annular mist (39).

Beyond blowout, in an experiment at constant velocity and gradually increasing solid rate, a group A powder does not choke but undergoes a gradual increase in solid fraction (58). The classic choking data for group B solids were obtained from a setup with a blower that surged (terminating the experiment) when choking imposed a sharp increase in the head that had to be overcome (59). Later data (60) reveal that a dense regime develops if the experiment is continued with a compressor able to give the higher head

(61). More work is needed to elucidate the triangle in Figs. 2 and 3, where transport and dense fluidization curves collide (41).

### Scale-up: Art or Science?

As one looks at the history of fluid beds, the low cost and speed of certain developments signal their intrinsic rightness. Little science entered into the development of Winkler's gasifier, the FCC (62), Albert Godel's Ignifluid boiler, Sasol's catalytic reactor, John Highley's AFBC with lumps of sized, washed coal floating on a shallow fluid bed of sand, Lurgi's CFB calciner, or the CFB boiler (63). Developing the bubbling AFBC with steam tubes within the fluid bed proved to be more difficult (64). Disappointments in early installations (65) taught that scale-up is largely a matter of taking care to provide for uniform gas flow into the bottom of the bed, uniform coal feed, adequate collection and recycling of carbonaceous fines, adequate freeboard height for combustion of fines, and protection of tubes from erosion and corrosion. But scientific study has helped. The literature provides mature descriptions of the hydrodynamic behavior of bubbling beds of group B solids (66), means for estimating residence times of carbon particles within a bubbling bed (30), and analysis of combustion of the particles (67).

Scale-up of group A fluid beds for catalysis is notoriously difficult when high conversions of reactants are desired. Figure 6A illustrates the usual progression from bench scale to commercial design. The lower curve in Fig. 6A suggests how this progression has sometimes led to a poor reactor efficiency (ratio of apparent kinetic constant to the intrinsic constant). In FCC regenerators, either poor designs or careless operators, who neglected to maintain an adequate level of fines, have led to reactor efficiencies as low as 10 percent (68). The Brownsville Fischer-Tropsch synthesis development, which skipped the demonstration plant stage of Fig. 6A, produced a reactor efficiency of about 2 percent (13). Gas bypassing in the form of large bubbles is the main culprit, although backmixing of gas (Fig. 6B) is a second negative factor. A third kinetic difficulty is the risk that catalyst will suffer harmful effects from its cyclic exposure to two quite different gases: bubble gas, containing unreacted species, and the highly converted gas of the emulsion phase (Fig. 6C). Exposure of catalyst particles to the first gas is brief,

whereas a particle's dwell time in the latter gas, in a poor design, can be relatively long. At Brownsville, emulsion gas oxidized iron catalyst to a noncatalytic oxide; catalyst from Brownsville quickly regained catalytic activity when, in laboratory beds, it was exposed for longer relative times to bubble gas. This is not an isolated warning of the unsteady-state nature of the kinetic scene that underlines the integral reaction kinetics of a fluid bed (69, 70). One should not expect, a priori, that these kinetics can be successfully modeled from differential reaction kinetics obtained for a solid exposed to a gas of constant composition. The unsteady-state nature of the kinetic scene in a fluid bed can sometimes be helpful (69, 71). Engineering opportunities for exploiting kinetic transients remain relatively unexplored.

Industrial designers of successful fluidization processes for group A powders have tended to ignore bubble theory and to focus on bubble suppression. In a bench-scale reactor, where bubble size is limited by the presence of the wall of the reactor (72), efficiencies can be as high as 75 percent. Bubbleless fluidization (73) can give comparable efficiencies and can be achieved by combinations of increased gas velocity (21, 74), controlled level of fines (21, 75) (Fig. 6D), increased ratio of bed height to diameter ( $L/D$ ), and staging. Increased gas velocity and fines level limit bubble life and size or move the operation into the turbulent or fast regime, reducing gas backmixing and relative dwell time of catalyst in converted gas. Staging (for example, with horizontal, perforated baffles) is particularly useful for slow reactions or when an adequate concentration of fines cannot be maintained.

Kinetic and thermodynamic considerations define process conditions, but the designer needs a good hydrodynamic description for modeling. An approach is to treat the fluid bed as a classical mass-transfer device (44, 76). Three parameters determine conversion: a coefficient for mass transfer between gas and catalyst, an axial gas-mixing coefficient (Peclet number), and a kinetic coefficient. The treatment is particularly suitable at desirable commercial conditions: sufficient fines, tall beds, and gas velocities in the turbulent or fast regime. Van Swaaij (77) has demonstrated the treatment in the successful scale-up of the Shell chlorine process. He calculated mass transfer and mixing coefficients from tracer experiments. Mobil Research and Development Corporation has successfully applied the treatment in scaling its methanol to gasoline (MTG)

process (78). As additional data on mass-transfer and mixing coefficients become available from experiments in large models, better correlations for these coefficients will eliminate the need for a costly demonstration plant.

### Methanol to Gasoline

In the early 1970's, Mobil scientists discovered that ZSM-5, a selective zeolite catalyst, converts methanol to high-octane gasoline (79). Bench-scale studies demonstrated the conversion in both fixed and fluid beds (80). The MTG reaction poses three difficulties to the reaction engineer. (i) It is highly exothermic; the heat of reaction, 1740 kJ per kilogram of methanol, would, if uncontrolled, produce an adiabatic temperature rise of 600°C. (ii) ZSM-5 catalyst deactivates both by irreversible steaming and reversible coke deposition. (iii) Conversion must be essentially complete, since unconverted methanol would be expensive to distill from water, a by-product of the MTG reaction. A fluid bed reactor offers obvious advantages in meeting the first two difficulties. Heat of reaction can be easily removed by circulating catalyst to a separate cooler, by placing heat-exchange tubes within the bed, or by feeding liquid methanol to the bed. Heat

removal in a fixed bed, either by indirectly cooling the bed or by injecting cold recycle gas in large amounts, is expensive. Continuous regeneration and a continuous supply of fresh makeup catalyst can maintain catalyst activity in a fluid bed at any desired level, but one must operate a fixed bed cyclically. The fluid bed is less expensive to build; its yield of gasoline is higher (92 percent compared to 85 percent from the fixed bed); and its gasoline has a higher octane number (96 Research Octane compared to 93 from the fixed bed).

After both the fluid bed and the fixed bed had operated at the 4 barrel per day scale [the fluid bed was 102 mm in internal diameter (81)], the New Zealand government inquired in 1978 about using MTG to convert natural gas to gasoline. Since New Zealand was unwilling to wait for a demonstration-scale fluid bed, prudence dictated the choice of fixed beds for the plant now nearing completion at Motunui, Taranaki, North Island. Beginning in January 1986, it will produce gasoline at 14,450 barrels per day (82).

Early 1985 saw completion of a 5-year international project successfully demonstrating MTG at 175 barrels per day in a 600-mm fluid bed (83), meeting the expected complete methanol conversion, high gasoline yield and quality, and low catalyst consumption.

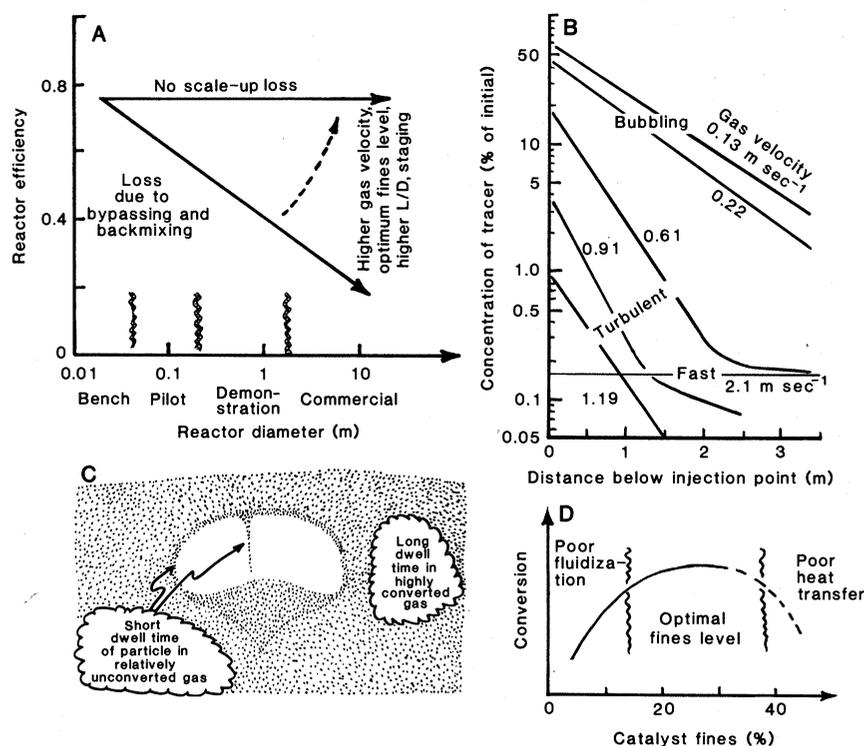


Fig. 6. (A) Variables affecting scale-up of a group A fluid bed reactor, roughly in order of importance. (B) Concentrations of a tracer gas at various distances below the point of injection into a group A powder in a 152-mm column (74). (C) The unsteady-state nature of the kinetic scene in a bubbling fluid bed (43, 69). (D) Effect of fines (<40  $\mu$ m) on performance of a group A powder (75).

## Conclusions

We expect the CFB boiler to be more attractive than the bubbling AFBC to utility engineers. We predict that the PC boiler will soon become obsolete for electricity generation. A supercharged CFB (1, 4) may win a role for retrofitting old stations, replacing old boilers and extending the life of old steam turbines, while reducing emissions of sulfur and nitrogen oxides.

The bubbling AFBC should find its role, probably in the multibed version, at steam rates too small for the CFB to be competitive. We suggest that about 30 to 60 MW (thermal) will be the range of energy production in which bubbling and CFB designs will compete, with bubbling beds winning most applications below 30 MW. A multibed version of the Ignifluid boiler (84), if someone were to develop it, might compete at far higher ratings, since the Ignifluid emits far coarser dust (which is much easier to control) than any of its competitors (85).

An urgent task is to study fast-regime gas backmixing for a group A powder in a larger column. Will backmixing be significantly greater than Fig. 6B indicates for a 152-mm column? This is an important question for scale-up when a desired product can react further to something of lesser or even negative value. If gas backmixing should prove to be excessive, installation of vertical surface in the fast regime may correct it.

In general, fluid beds are more competitive in larger sizes (86), and reaction engineers may always prefer the fixed-bed reactor for small-scale applications. But an objective of fluidization research should be to enable a large project like New Zealand's to consider the fluid bed without waiting for results from a demonstration plant. Data from the large number of cold models built recently to study group A powders at high velocities (50) should soon provide better correlations for coefficients of mass transfer and axial mixing in the turbulent and fast regimes. We urge experimenters to keep the level of fines in view as an explicit variable. When the correlations appear, the large fixed-bed catalytic reactor may follow the PC boiler into disuse.

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6. Designs with two stages of combustion, the first being substoichiometric, give the lowest nitrogen oxide emissions (for example, II, V, and VII in Fig. 2 and VI in Fig. 3).
7. Sparked by Henrik Harboe, Stal-Laval Turbin has worked from the early 1970's on pressurized fluid bed combustion (PFBC) to be used in combination with gas- and steam-turbine power cycles. Not a member of EPRI, American Electric Power (AEP) has collaborated with Stal-Laval Turbin and Deutsche Babcock in a 15-MW PFBC experiment at Malmö. ASEA has announced a project to build two PFBC units in Stockholm for a total of 131 MW (electric). AEP has proposed a 70-MW PFBC demonstration at its Tidd station, near Steubenville, Ohio.
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