Chapter 1
Introduction

Abstract This introductory chapter begins with a description of the various flow regimes observed in beds of solid particles fluidized with gases or liquids. Distinctions are drawn between the gas-fluidized behaviour of fine, low density particles, Group A in the Geldart classification, larger, more dense materials, Group B, and those more difficult to fluidize at all, Groups C and D. It is shown how gas-fluidized beds behave as the velocity of the gas flowing through them is increased the transitions from bubbling to slugging to turbulent to transport behaviour being described and illustrated by means of schematic diagrams. There then follows a discussion of the historical development of fluidization from its first major industrial application in the 1940s through subsequent decades up to the first decade of the new millennium. Theoretical advances are described from the early “two-phase” theory of Toomey and Johnstone through Davidson’s analysis of the flow of gas through bubbling beds to the more recent “particle-bed” model. Industrial applications of gas-fluidized-bed reactors are discussed with reference to fluidized catalytic cracking, naphthalene oxidation, coal to gasoline via the Synthol process, propylene ammoxidation to acrylonitrile, ethylene polymerisation and butane oxidation to maleic anhydride. The wide range of experimental techniques used in basic fluidization research is described as well as the recent application of computational fluid dynamics to gas-solid fluidized systems.

1.1 Regimes of Fluidization

Two-phase systems: liquid-solid; gas-solid
An assembly of solid particles becomes fluidized when the combined drag and body forces exerted on the particles by an upward-flowing fluid exceed the gravitational force holding the assembly together. At this point the particles are suspended in the fluid and are free to move. The fluid velocity at which this occurs is termed the “minimum fluidization velocity” $U_{mf}$ and its value is a function of the size, shape and density of the particles and the density and viscosity of the fluid. When the velocity
of the fluid is increased above $U_{mf}$ the behaviour of the assembly, known as a “fluidized bed”, depends on the nature of both fluid and particle. If the fluid is a liquid the bed of particles expands more or less uniformly the particles moving about freely in the fluid flow field and stabilising at a void fraction, $\varepsilon$, which is a function of the fluid velocity, $U$, the terminal fall velocity of the particles in the fluid, $U_t$, and an exponent, $n$, which is a function of the fluid-particle system in question; $n$ correlates with the terminal Reynolds number, $Re_t$, but is constant in the viscous and inertial flow regimes at 4.8 and 2.4 respectively. Thus:

$$\frac{U}{U_t} = \varepsilon^n$$

Equation (1.1) is known as the Richardson-Zaki equation and applies to a wide range of particle-liquid combinations (see below).

If the fluidizing fluid is a gas the behaviour of the bed as $U$ exceeds $U_{mf}$ depends on the nature of the particles. Geldart (1973) classified fluidizable particles into four groups, A, B, C and D as shown in Fig. 1.1.

As $U_{mf}$ is exceeded particles in Group A expand uniformly in the same way as a liquid-fluidized bed up to a gas velocity at which the bed collapses and voids called “bubbles” begin to form; these rise through the bed at a velocity proportional to their size and burst at the bed surface. The gas velocity at which this occurs is termed the “minimum bubbling velocity, $U_{mb}$”. A typical example of a Group A powder is cracking catalyst used in the FCC process to be described later. Group B particles such as coarse sand begin to form bubbles immediately $U$ exceeds $U_{mf}$ a type of behaviour referred to as “aggregative fluidization”. The particles in Group C, typically flour and cement, tend to be cohesive and are difficult to fluidize at all whereas the relatively large and dense materials in Group D such as lead shot are prone to spouting rather than fluidizing. The great majority of fluidized-bed reactors use powders in Groups A and B.

![Fig. 1.1 The Geldart classification of powders](image-url)
Once the bubbling regime has been established in beds of A and B materials any increase in volumetric gas flow rate causes the bubbles to coalesce, grow in size and increase in velocity as they pass to the bed surface. With increasing superficial gas velocity in beds of material contained in a vessel with a high aspect ratio (height to diameter ratio) a point is reached at which the bubble diameter becomes equal to that of the container and the bed is then said to be “slugging” and is characterised by the periodic passing of large bubbles or slugs and large fluctuations in bed pressure drop corresponding to the slug frequency.

In bubbling beds of wide diameter particles are ejected into the freeboard space above the bed surface by the bursting bubbles the amount of material entrained increasing as the gas velocity is increased. At sufficiently high gas velocities bubble flow breaks down, the bed becomes highly turbulent and entrained particles must be separated from the gas flow via a cyclone separator and returned to the bed. The transition from bubbling to turbulent has been characterised by a critical gas velocity, \( U_c \), at which the standard deviation of pressure fluctuations measured at the base of the bed reaches a maximum value and axial and radial solids density profiles are more or less uniform over time. Operating gas velocities in the turbulent regime are in the range 0.4–1.2 m s\(^{-1}\). A further transition from turbulent to so-called “fast” fluidization occurs at gas velocities in the range 1.2–15 m s\(^{-1}\). At these velocities considerable entrainment of particles occurs and the bed develops a lean-core-annulus structure with a parabolic radial solids density profile. Solids are observed to move downwards at the walls of the containing vessel in a refluxing type motion. To maintain steady-state operation solids circulation via an external standpipe is necessary and as a result the beds are often turbulent in the base region and fast at the top with a pronounced axial solids density profile. These systems are normally referred to as circulating fluidized beds or CFB’s.

A schematic diagram of the various regimes of fluidization is shown in Fig. 1.2 and a detailed discussion of much of the work carried out to distinguish between them is given by Grace and Bi (1997).

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**Fig. 1.2** Flow patterns in gas-solid fluidized beds (Grace and Bi 1997)
Three-phase systems: gas-liquid-solid
In these systems solid particles in a column are held in suspension, i.e. fluidized, by an up-flowing liquid, a gas being introduced separately at the base of the column. Thus liquid and gas flow co-current upwards the liquid being the continuous phase and the gas the discontinuous, bubbling phase. Several different modes of operation are possible: the solids may be retained in the column as in the case of a conventional liquid-solid fluidized bed or they may be transported out at the top and reintroduced at the base as in the case of a fast gas-solid system. The hydrodynamics of three-phase fluidized beds are complex but have been reviewed comprehensively by Fan (1989, 2003). Three-phase systems have been applied in a number of important industrial processes such as the catalytic hydrogenation of heavy or residual oils, the Fischer-Tropsch process, fermentation and aerobic biological waste water treatment.

1.2 A Brief History of Fluidization

The following is a brief outline of the major highlights in the story of fluidization tracing the development of the subject from its emergence in the early 1940s to the present day and dividing the period up into decades. Like centuries decades are arbitrary divisions of time but they represent a convenient way of charting industrial and academic developments in this area. Furthermore there is no doubt that the period 1940–1950 was decisive in the story of fluidization.

1.2.1 The 1940s

1.2.1.1 Fluidized Catalytic Cracking

On 25 May 1942 the world’s first commercial fluidized catalytic cracking unit began operation at the Baton Rouge refinery of the Standard Oil Company of New Jersey (Jahnig et al. 1980). There had been two industrial uses of fluidization prior to this but neither had been very successful and they were applied on a very limited scale. The fluidized catalytic cracking, or FCC, process was the first major application of the technique and its commercialisation was the result of an enormous research and development effort by oil companies, plant contractors and academics. The subsequent spread of the process to virtually every major oil refinery in the world is a measure of the success of that effort.

The driving force that led to the development of the process was the need for a gasoline of higher quality than had been available hitherto for the rapidly improving internal combustion engines that were coming onto the market in the 1920s and
Gasoline had been produced since the earliest days of oil refining first as a straight-run distillate fraction and later by thermal cracking of heavier fractions such as gas oil. The quality of gasoline produced by thermal cracking was sufficiently superior to the straight-run variety as to make the process a viable proposition. The realisation of the link between the structure of the gasoline molecules and their performance in engines only came about in the 1920s and this led to the introduction of the “octane number” scale on which the linear n-heptane was given an octane number of zero and the branched iso-octane a number of 100. The performance of a gasoline was then compared with blends of these two materials in a standard test engine to determine the octane number of the fuel. The product from thermal cracking was more highly branched than the straight-run material and this accounted for its superior performance. Now it might be thought that a suitable catalyst could have been found that would increase the extent of branching even further. Catalytic processes had for long been used in the chemical industry for just such purposes but oil companies at that time were reluctant to become involved with catalytic chemistry. They preferred to keep to physical operations such as distillation or simple chemistry such as thermal decomposition with which they were familiar. The catalytic effect of aluminium trichloride on the cracking of oil fractions had in fact been observed in 1915 but it wasn’t until the pioneering work of the French mechanical engineer Eugene Houdry in the mid-1930s that a catalytic cracking process became a reality. He discovered through trial and error experiments that a naturally occurring clay mineral that had been activated by treatment with acids was capable of carrying out the cracking reaction and of producing a highly branched material with a superior performance to the thermally cracked product.

Houdry discovered that in addition to producing gasoline his catalyst also produced a form of carbon called “coke” which was deposited on the catalyst surface and gradually reduced its activity to zero. He found however that the coke could be burned off by blowing air over the surface at about 500 °C. On the plant this was achieved by having three packed-bed reactors on stream simultaneously, one cracking, one regenerating and one being purged ready for the next stage. The whole cycle took 24 min and its success was strongly dependent on the use of high-temperature automatic valves and sophisticated control algorithms both of which were revolutionary at the time.

The evident success of the Houdry process caused the world’s biggest oil company, Standard Oil New Jersey (now ExxonMobil), to become more interested in catalytic cracking than they had been hitherto so in early 1938 with a European war on the horizon and an anticipated need for large quantities of aviation gasoline the company took the decision to develop a catalytic cracking process that would be independent of Houdry’s patents. A consortium, Catalytic Research Associates, was set up consisting of eight companies: Jersey Standard, Kellogg, Indiana Standard, Anglo Iranian (which later became BP), Universal Oil Products, Texaco, Shell and the German chemical giant I G Farben (which dropped out in 1940). As one observer has commented “This was a formidable grouping and, with 400 workers at Jersey and 600 in the other companies, represented probably the largest single
Fluidized-Bed Reactors: Processes and Operating Conditions
Yates, J.G.; Lettieri, P.
2016, XVIII, 205 p. 66 illus., 7 illus. in color., Hardcover
ISBN: 978-3-319-39591-3