Fluidized Bed Technology: Challenges and Perspectives

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Abstract. Fluidized beds are recognized as important gas-solid contacting technology in chemical synthesis reactors, in combustion/gasification/pyrolysis of coal and biomass, in multiple drying applications, in solar thermal energy capture and storage, and more recently also in the production of hydrogen by either catalytic steam reforming of organic feedstock, or thermal water splitting using oxidation-reduction cycles. Although the process understanding and operational experience has significantly been expanded over the past decades, some challenges remain the be dealt with, as discussed in the paper. These challenges are mostly of gas-solid hydrodynamic nature.

1. Industrial Background

Despite its first commercial use as in 1926 for the gasification of coal, its widespread application started in the early 1940s with the construction of the first fluid bed catalytic cracker (FCC) by Standard Oil of New Jersey. The first commercial unit went into operation in May 1942 and was the culmination of an extensive scale-up procedure from small laboratory units via a semi-commercial 100 barrel per day plant, and several years of research on methods for contacting gas with a fine solid catalyst. Professors Lewis and Gilliland of MIT, who were consultants on the project, considered several options including dilute-phase pneumatic conveying, the “down-flow design” (or bubbling bed as we now know it) and the “up flow design” (or fast fluid bed as it is now known) (see Figure 1). The bubbling bed was rejected at first because the low gas velocities (<0.5 m/s) implied vessels having a large diameter; the dilute phase flow regime required very tall vessels or the use of multiple tube coiled reactors, both of which gave temperature gradients, and this approach was also rejected. The first choice was the upflow (fast bed) design operating at about 2.5 m/s and the unit put on steam in 1942 had a reactor 4.5 diameter and a regenerator 5.9 m diameter. In their patent Lewis and Gilliland (1950) describe the operating principle of an upflow reactor as a concept whereby the reactor is operated at a fairly high superficial gas velocity with a continuous carry-over of the particles, but where these particles are collected and recycled to the reactor to maintain the bed inventory at a high concentration of particles.
Several upflow design plants were in operation by the end of 1943 but they experienced difficulties in solid separation, cyclone and standpipe design. It was decided to switch to the “down flow” or bubbling bed design in which there was a solids/gas interface which could be controlled and in which the low velocities (<0.6 m/s) gave much reduced particle entrainment. Over the years catalysts have improved in activity and selectivity with the result that contact times required in the reactor are much less than before. This has meant that gas velocities in the reactors could be increased without the need of a corresponding increase in vessel height; thus many cat crackers operate with an upflow or transport line reactor and a bubbling bed regenerator [1–4].

Encouraged by the success of cat cracking of oil, other fluid bed processes were developed in which the bubbling bed regime was used. Acrylonitrile manufacture is one such process. Propylene, air and ammonia are reacted exothermically in the presence of a catalyst similar in physical properties to FCC. The oxychlorination of ethylene, using air (or oxygen), and HCl to give vinyl chloride is another similar process [5,6]. Worldwide, there are at least 500 large fluid bed units operating these three processes, 350 of which are crackers.

All three processes use catalysts having a mean particle size in the range 40 – 75 μm and a particle density 1000 – 1800 kg/m3. The mean size, density, and the level of “fines”, usually characterized by the weight percentage less than 45 μm, are all known to influence both the flow properties of the catalyst and the chemical conversion. The sphericity of the particles is also thought to be important. However, in spite of the fact that these processes have been operated for 30 – 40 years, we still do not understand with any certainty the mechanisms by which the parameters listed above influence the behavior of these very important pieces of chemical plant. Also novel applications of group A particles are currently being developed, mostly in the bubbling fluidized bed operating mode, and targeting new fields of use [7–13].

An understanding of gas/solid interactions, particularly of very fine particle systems, can open the way to new processes and products in the 21st century, provided adequate solutions are found for the remaining technical, design and operational challenges which these fluidized bed applications present.
2. Technical background

2.1. Powder properties

It is now widely accepted that powders can be divided broadly into four-man groups according to their fluidization/aerated flow characteristics. These groups were introduced by Geldart [14] as Cohesive, Aeratable, Bubbling and Different (coarse) particle classes. A distinction between these particle groups can be made on the basis of the \( d \) and absolute particle density, \( \rho_p \). Kong et al. [9,15] delineated the particle groups. The C to A boundary can be represented by Eq. 1, with \( d \) in \( \mu \text{m} \).

\[
d_{C\rightarrow A} = \frac{10325}{(\rho_s - \rho_g)^{0.725}}
\]

The A to B powder transition follows

\[
d_{A\rightarrow B} = \frac{907200}{(\rho_s - \rho_g)^{1.17}}
\]

B-class powders move into the D-class group at

\[
d_{B\rightarrow D} = \frac{266300}{(\rho_s - \rho_g)^{0.807}}
\]

Our main interest is into the fine particles, i.e. those in groups A and C. FCC, acrylonitrile and oxychlorination catalyst are located right in the middle of group A, but the powder collected in the second and third stage cyclones is more likely to be in the cohesive group C. Beds of powder in group A expand considerably at velocities between minimum fluidization, \( U_{mf} \), and the velocity at which bubbling commences, \( U_{mb} \). Above the minimum bubbling velocity bubbles form, and grow rapidly with distance above the distributor, and with increasing gas velocity however to a maximum bubble size [16,17]. It is the size of these bubbles and the contact between the gas in the bubbles and the catalyst which determines chemical conversion. The flowability and extensive solids circulation within a bed of group A powders makes them easily conveyed around particle conveying loops. The ease with which they become and remain aerated however can provoke flooding on discharge from hoppers.

In group C, powders are cohesive and difficult to fluidize. Because the mass of each particle is small, the interparticle forces are comparable with, or greater than, the gravitational forces. These interparticle forces (electrostatic, Van der waals, vapour and liquid adsorption) are dependent on the size, shape, roughness, hardness and chemical structure of the particles as well as on the gas properties.

2.2. Plant Design and Operational Problems

What then are the problems in these catalytic processes, how are they influenced by the particle and gas properties, and why have they not been completely solved after these many years?

A simplified FCC unit is depicted in Figure 2.

There are many variations on the basic design but essentially the engineering and economic success of a catalytic cracker depends on (i) the ability to circulate solids around the system at a controllable rate since this determines the heat balance and therefore the throughput; and (ii) the ability to keep catalyst losses down to an acceptable environmental and economic level.
Whereas the original FCC unit lost on average approximately 5 tons catalyst per day at a cost of £1000 per ton, 7 days per week, 52 weeks per year, new de-dusting equipment [18] has reduced this considerably. These losses routinely arise largely from attrition of the catalyst [19]. The hardness of the catalyst has a direct bearing on these losses and the response of the catalyst manufacturers has been to make catalysts harder. In doing so the catalysts have been made denser, and this has affected adversely the ability to circulate the solids. Two effects occur, mostly in the diplegs (Figure 3) [20,21].

(i) The powder de-aerates more quickly. Thus, in flowing down tall standpipes where the build-up in hydrostatic pressure causes the gas volume to decrease, the powder may become de-fluidized.

(ii) The maximum stable bubble size increases, large bubbles form which occupy a large proportion of the standpipe cross section and choke off the downwards flow of solids.
Occasionally large losses of catalyst can occur through malfunction of the catalyst dipleg and reduce the cyclone efficiency, again causing losses.

The Acrylonitrile and oxychlorination reactors are illustrated in Figure 4 [5]. These two processes use fluid beds which, superficially, look very similar to each other. The reactors contain a large number of vertical heat exchange tubes and the reactions involve air, a hydrocarbon (propylene) and ammonia in the former case; in the vinyl chloride monomer reaction, ethylene and hydrochloric acid are fed to the reactor. In both cases complete premixing of all three components is impossible because of possible explosions, so the gases must be brought into intimate contact with each other and the catalyst inside the reactor. Not only is gas distribution important [22], but so also is the size of the bubbles, entrainment of solids and their return to the bed through primary, secondary and tertiary cyclones. Catalyst losses through attrition and successful operation of diplegs and trickle valves also play a role in the successful operation of both processes.

**Figure 4.** Oxychlorination reactor

**Figure 5.** Bubble splitting

### 3. The Technical Challenges

Having posed the problems, we must ask two questions: “how are they influenced by the particle and gas properties, and why?” An understanding of the fundamentals – the “why” – is essential to producing long term solutions.
Empirical observations have shown that, providing the catalyst does not become cohesive, reducing mean particle size, reducing particle density and increasing the mass fraction less than 45 µm improves catalyst circulation. The powder is less likely to become de-fluidized due to gas compression and maximum stable bubble sizes are reduced. An increase in gas pressure and gas viscosity also improves the powder behavior and can be described by empirical equations but as yet there is no satisfactory theoretical explanation why. The concept of a maximum stable bubble size implies not those bubbles reach a certain size and then remain unchanged but rather that a state of dynamic equilibrium is reached in which splitting and coalescence occur continually as shown (Figure 5). The mode of bubble splitting is assumed to be either (i) by formation of an indentation which forms on the upper surface of the bubble and grows as it is swept around the periphery while growing to reach the floor of the bubble before being swept away until the bubble will split; or (ii) by a penetration of particles from bubble wake up to the bubble roof, mostly occurring during bubble coalescence.

Also the bed temperature seems to significantly affect the maximum stable bubble size, with a significant reduction as the temperature of operation increases [9,15]. The underlying mechanisms are yet not fully elucidated. Do the interparticle forces or hydrodynamic forces cause the changes in the dense phase? If the former, then which interparticle forces are responsible and how are these affected by pressure and temperature? Certainly, further experiments are required to verify this.

What are the hydrodynamic conditions inside a cyclone dipleg, and how does a trickle valve operate? How do the powder properties affect the behavior and does it matter if additional purge gas is added to the bottom of the valve?

Surprisingly these questions had not yet been studied systematically. Some surprising and fascinating answers are being found. Research has indeed produced evidence that quite small percentages (<5%) of very fine (<10 µm) particles can give large increases in bed expansion and de-aeration times. These very fine particles are not removed quickly by entrainment from the fluidized bed, as one might except; one the contrary their entrainment rate is a factor of ten less than predicted by correlations based on large particles [23,24]. Here again, interparticle forces seem to be very important.

Other work has demonstrated that the sphericity of the particles affects the bubble size in fluidized beds of group A particles, with bubbles in beds of spherical particles shown to be significantly smaller than those observed when the particles were angular. Why should particle shape influence bubble size?

In many areas of technology, both mathematical and experimental modeling, can be done to simulate real situations using dimensionless groups. Examples of this are to be found in flow through pipes (Reynolds numbers), ship and aircraft design (Reynolds and Froude numbers), heat transfer (Nusselt and Prandtl numbers) and many others. Although many of these situations are complex, the basic physics is understood so that the important dimensionless groups can be derived and then used for scaling. In fine particle aerated flow, the basic physics is not understood and we are driven to develop empirical correlations based often on small cold atmospheric pressure models, and use them under pressurized and/or hot large units.

And improvement of less than 0.5% conversion in any one of the three processes discussed earlier can result in an increase in profitability measured in millions of US $ worldwide. So why is there not more research being done? Most experiments carried out in the laboratory cannot simulate even approximately the conditions on a large plant, and it would be a courageous and foolish consultant and/or process engineer who would guarantee that any given modification would be successful, and would not result in other unforeseen and undesirable consequences. The problem is that it can be disastrously expensive to introduce changes on a large plant if the “improvement” doesn’t work. Most plant managers agree with one of Murphy’s laws “If something is working well enough, don’t try to fix it”.

Over the past decade modifications intended to improve the performance of one FCC unit cost well over a million US $ in down time and lost production when they didn’t work; improvements on an oxychlorination reactor cost a similar sum for the same reason.

Nevertheless, evolutionary improvements are being made but they take a long time. There are many challenging problems and their solution demands both fundamental research into basic mechanisms as well as developmental work on a pilot plant scale.
The newest challenge in fluidized bed technology has come with the increased application of the circulating fluidized bed – essentially the “up-flow operation” shown in Figure 6 – to fluidized bed combustion and other processes.

There is a lot of speculation about the behavior of the gas and the particles inside the device, largely due to observations made in small, usually two-dimensional units. Claims are still being made, though less vigorously, that solids distribution is relatively uniform across the diameter of the vessels, and that the reason why the particles remain in the vessel even though the gas velocity, is that the particles cluster together as large, loose agglomerates. The alternative view is that the CFB consists of a relatively dilute up flowing core surrounded by denser down flowing suspended solids at the wall. These two perspectives have very different implications for scale-up strategy. The cluster theory supports the idea of relatively trouble-free scale-up whilst the core-wall approach suggests problems with increasing scale. The biggest CFB combustors now commissioned have diameters up to 10 m diameter and the consequences of having incomplete combustion because of poor radial mixing will be very serious. However, it is questionable whether research could have resolved this in advance. Sometimes you just have to take a chance.

![Figure 6. Typical circulating fluid bed combustor](image)

4. Conclusions

Although the process understanding and operational experience has significantly been expanded over the past decades, some challenges remain to be dealt with, as discussed in the paper. These challenges are mostly of gas-solid hydrodynamic nature. Certainly, Computational Fluid Dynamics applications offer a powerful tool in assessing this behavior and are currently widely applied in the design and operation stages of the fluidized bed processes. Fluidization will keep researchers and designers busy for many years to come.
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Acknowledgments
The research was supported by The Beijing Advanced Innovation Centre of Smart Matter Science and Engineering of the Beijing University of Chemical Technology, Beijing, China.