Fluid dynamic regimes in circulating fluidized bed boilers—A mini-review

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HIGHLIGHTS

- Circulating fluidized bed boilers are usually operated with a bubbling bottom bed.
- The upper furnace in circulating fluidized bed boilers is not fast fluidized.
- The fluidization regimes in boilers should be better defined.

1. Introduction

1.1. General

There is a great number of publications on the regimes of fluidization in circulating fluidized bed (CFB) systems. These investigations can be divided into those related to the phenomenon, characterizing a regime of fluidization, and those describing regimes in a particular device, such as in the axial positions of a CFB.

Sun and Zhu (2018) present examples of published regime maps as an introduction to their own work on the same theme. One of the most influential representations is that of Grace (1986), later completed and explained, for example by Bi and Grace (1995). Grace's regime map has been included in literature surveys and textbooks, for example by Kunii and Levenspiel (1991), and it is often referred to. In this map the circulating fluidized bed is located at higher fluidization velocities than those of bubbling beds. In the version of Kunii and Levenspiel the denomination “Circulating beds” in the original diagram is replaced by “Fast fluidization”. This seems consistent, since fluidization phenomena are described rather than what occurs in beds related to a certain reactor. Because of the focus on phenomena there is no distinction between different parts in a reactor, such as bottom bed or splash zone, although the authors sometimes refer to the “S-shape” density profile of Li and Kwauk (1994), which comprises a dense bottom bed coexisting with a disperse top region in a riser. The latter authors covered a range of options, but their interest is in catalytic Group A particles, where in the case of the S-shaped profile, the bottom bed is fast fluidized and the upper part is in dilute-phase flow. They also point out that the transition between turbulent and fast fluidization may be difficult to perceive. The view on phenomena is further emphasized by the listing of sequential regime transitions as a function of velocity given by, for instance,
Bi and Grace (1995). For Group B particles, the transport version of this sequence would be dense phase flow, bubbly flow, slug flow, turbulent flow including fast fluidization, core-annular dilute-phase flow, homogeneous dilute-phase flow, where the fast flow is delimited by classical and accumulative choking.

Most of the knowledge underlying the picture of fluidization regimes, presented in the publications mentioned, was derived from investigations in narrow laboratory risers aiming at various chemical engineering applications, particularly fluidized catalytic crackers, but implicitly also dealing with other applications, such as boilers. However, CFB boilers are wide and tall and operated with beds formed predominantly by ashes from the fuels burnt, resulting in a wide distribution of particle size. So, there is a difference in the conditions applied in the tests on fluidization regimes and in the regimes in boilers (vessel size, particle size distribution, temperature). Actually, particle size distributions have been investigated by Sun and Grace (1992) but size distributions are further-more related to the CFB system in the form of segregation, which has been mentioned in a few of the references quoted below (Yue (2017) and Wang et al. (2021)). The combustion chambers of large commercial boilers are difficult to access for measurements because of their size and because they are covered by membrane-tube walls. Occasional service openings in boiler furnaces may not be in representative positions or have a suitable form for investigations. Consequently, few measurements are available from such units, and systematic parameter variations are lacking. Therefore, information from smaller units, forming the basis of what was resumed above, is the knowledge base available for judging the fluidization regimes in boilers.

1.2. Boiler features

The regime maps are intended to be generally valid, which implies that they should also be valid for boilers. A description that is more directly aiming at CFB boilers was published by Basu (2015) in his monograph “CFB boilers”. Basu uses the available information to assess the fluidization regimes in boilers. A chapter dedicated to fluidization regimes illustrates the uncertainty in dealing with this topic. The author, although aware of the possibility of a turbulent or a bubbling bottom bed, prefers to treat the bed as “fast fluidized” in figures and tables with an unspecified denser region below the secondary air injection level.

A common opinion about the fluid-dynamic regimes in CFB boilers is that there is a bottom bed operating under turbulent conditions and that the upper zone of the furnace contains a fast bed, or, as sometimes loosely assumed, that the entire bed is fast fluidized. These concepts on the state of fluidization require rigorous definitions to make a meaningful discussion possible on the conditions prevailing in boilers.

The turbulent bed has nothing to do with turbulence in the common fluid-dynamic sense, but it is a concept given to a fluidized bed, which is distinctly different from a bubbling or slugging bed because of its small, rapidly formed and disappearing voids, giving the bed a stirred appearance. This state can be determined by measurements of the amplitude of the pressure fluctuations in a bed. When the gas velocity in a fluidized bed increases, the bubble (or slug) size increases, and so does the amplitude of the pressure fluctuations. At a certain velocity \(u_r\), the amplitude does not increase further during a continued increase in velocity, and the character of the bed changes into the turbulent mode of fluidization while the amplitude of the fluctuation decreases. When the velocity increases further after a transition period, the bed gradually starts circulating, denoted by the transport velocity \(u_t\). Both velocities are determined empirically as functions of the Archimedes number \(Ar\), that is, the bed particle size has a significant influence, Bi et al. (2000) presents an overview. So, here is already a contradiction: by definition, the bed starts circulating at \(u_r\), but the bottom bed of a CFB is sometimes claimed to be operated under turbulent conditions, which, according to its definition is non-circulating. A benevolent interpretation is that the particles in the (turbulent) bottom bed are coarser than the circulating particles, which are smaller and then subject to transport conditions. However, this is usually not mentioned in publications. In contrast to the general opinion, just related, Leckner (2017) and coworkers repeatedly claimed to have observed a bubbling bottom bed in a CFB boiler operated at high velocity. Werther (1993) also adapted this idea and explicitly referred to the work of Werther and Wein (1994) in his conclusion that the bottom bed is bubbling. Despite that, in the paper of Werther and Wein (1994) it was stated that they investigated a turbulent bed under CFB conditions. In a recent review of literature, Wang et al. (2021) found that the bottom bed of a CFB boiler is bubbling. Yue et al. (2017) also mention a bubbling bottom bed without proof or further comment, except that the bed quality may be characterized by coarse ash particles because of the coal quality used. In two regime maps, Squires et al. (1985) clearly classify CFB boilers as having a turbulent bottom bed. (Sekret and Nowak, 2006) measured the amplitude of the pressure fluctuations in a 670 MW, CFB boiler and found a maximum, indicating a turbulent bed. They called it “bubbling-turbulent”. However, this maximum can be explained by the rise in bubble size with increasing velocity until the bubble size was in the order of the bottom bed height. At higher velocities, the decline in the amplitude could be explained by the decline in bottom-bed height during the increase of velocity, because of redistribution of bed material from the bottom bed to other parts of the recirculating loop. Hence, the bed was still bubbling and not turbulent. In the dominant patent on CFB boilers (Reh et al. 1979) it is written “There is no drastic change in density between a dense phase and an overlying dust-containing space but the concentration of solids in the reactor decreases continuously in an upward direction to the place at which the solids are entrained out of the reactor in a gas stream”. This reminds of the “S-shaped” profile for Group A particles investigated by Kwauk and co-workers. The fluidization regime was not mentioned, though, but the lack of a “drastic change” in density made the bottom bed in this patent different from a bubbling bed, which does have such a change, and the bed was obviously fast fluidized or turbulent. This distinction was recognized legally and obliged other manufacturers of CFB boilers to have a license agreement with Lurgi, the patent owner. The existence of patent licenses is confirmed in the review of Goral et al. (2017), mentioning a general transfer of knowledge about CFB technology from Lurgi to other CFB manufacturers. As an exception, Foster Wheeler developed their own process characterized by the presence of a pronounced bed in the bottom of the furnace and a relatively solids-lean freeboard above it. They mention “An alternative process, i.e., fast fluidized or highly expanded bed, is characterized by having the solids spread over a substantial height of the furnace,” Abdulally et al. (1992). The “alternative process” coincides with that described in the Lurgi patent. (Unintend-edly Foster Wheeler obtained the license when they purchased Alstom-Pyropower in 1995, but the patent expired some years later).

1.3. The dilute region

In the upper part of the fluidized bed riser (the furnace in a boiler) there is also some ambiguity in definitions. The fluidization regime in this part, in fact often including the entire bed, is called “fast fluidization”, following Verushalmi et al. (1976) who worked with fine-particle reactors. It is an attractive denomination as a contrast to a bubbling bed, called a “slow bed” by Verushalmi (1978). However, the problem is that fast fluidization is used to
denote a fluidization regime. For such a concept to be valid, a strict definition is required, and a generally accepted description is not available for two-phase flow in boiler furnaces. Particularly uncertain is the range of validity of the concept of fast fluidization in the most dilute region (at low particle concentration), found in boilers, compared with regime maps (for instance, those of Li and Kwaak 1980; Yerushalmi et al. 1985; Bai et al. 1993). This will be further discussed below. A definition is necessary, because this concept is related to certain properties of the flow, such as clustering and other phenomena. It should be mentioned that this topic has been treated recently by Sun and Zhu (2018) and by references in that publication.

2. The bottom bed

Based on measurements of pressure fluctuations Leckner (2017) claims to have observed a dense bubbling bed in the bottom zone of the Chalmers 12 MWt boiler during regular operation conditions. First, it should be explained that this boiler is well equipped with pressure taps from the bottom to the top of the furnace. Fig. 1 shows an example of pressure measurements from normal CFB operation.

The bottom zone, Zone 1, shows the linear pressure drop with height, typically observed in a dense bubbling bed. This zone is followed by a transition zone, Zone 2, normally called “splash zone”, again a characteristic of a bubbling bed where the bed material is ejected from the bottom bed by bursting bubbles. If the total amount of bed material in the system is too small, the bottom zone disappears and turns into a smaller transition zone while the bed is distributed in the CFB system. Then, also the extended splash zone disappears. However, usually CFB boilers are operated with bottom beds. Similar observations of the decay of pressure with height are rarely available in publications concerning the pressure development in CFB risers. The reason is that, normally, very few pressure taps are installed, in contrast to those of the Chalmers boiler. With few pressure taps, the determination of the behaviour of the narrow bottom zone becomes uncertain, and so are the interpretations of the state of the bottom bed in CFB risers.

Bubbles are shown to exist even at higher fluidization velocities than the terminal velocity of single particles. The reason, as claimed by Leckner (2017), is that the particle phase in the bottom bed does not experience an excessively high velocity because a great deal of the gas passes as through-flow through the bubbles. This fact has not been measured in a CFB bottom bed, operating at high velocities, but is based on visual observations through the transparent walls in down-scaled cold beds and on extrapolation of the knowledge from the flow characteristics of bubbling beds, which have an increasing through-flow at increasing fluidization velocity, such as shown in Fig. 2. The figure illustrates the increasing through-flow passing a bubble as the fluidization velocity increases in a bubbling bed, calculated according to a fluidization model.

One could claim that the linear fall-off of pressure with height in the bottom part, seen in Fig. 1, could have been produced as well by a turbulent bed. However, if the bottom bed were turbulent, the splash zone would not have been so developed as seen in Fig. 1. A strong indication on the persistence of the bubbling mode of fluidization is that the amplitude of the pressure fluctuations does not go through a maximum while the velocity increases, Leckner (2017). Although the high-velocity bed has the character of a bubbling bed and the concepts of “bubbles” and “emulsion phase” is used, it should be emphasised that the bubbles are not similar to those observed in calmly fluidized beds at low velocity. They may be denoted as "voids". The lower left image of Fig. 3 gives an idea. Because of this gradually more irregular form of the bubbles and the lumps of particles forming the particle phase with the increase of velocity, the conventional bubble model used to produce Fig. 2 exceeds its range of confirmed validity. The extrapolated trends, however, should be valid for a qualitative judgement and that is sufficient to support the idea about the increasing through-flow with velocity.

In a review on gas–solid turbulent fluidization, Bi et al. (2000) divided turbulent beds into two classes, Type I, which occurs with Group A particles in non-slugging systems, where the maximum stable bubble diameter, attained at increasing velocity, is essen-

![Fig. 1. Time-average pressures, measured along the height of the furnace wall of the 12 MWt boiler at Chalmers University at a fluidization velocity of around 6 m/s with a sand bed of about 0.5 mm particle size, only insignificantly diluted with fuel ashes. The figure shows the bottom bed (1), the splash zone (2), the transport zone (3), and the top or exit zone (4). The pressures are recorded by individual pressure transducers mounted on inclined pressure taps, ending flush with the furnace wall. The pressure drop due to acceleration was estimated to 30 Pa, Johnsson and Leckner (1995).](image1)

![Fig. 2. Typical flow division in a bubbling fluidized bed, expressed in terms of fluidization velocities. The diagram distinguishes between through-flow ($U_{tf}$), visible bubble flow ($U_{vis}$), and flow through the particle phase ($U_{mf}$) as obtained from model calculations, Johnsson et al. (1991).](image2)
In a recent survey, Wang et al. (2021) also concluded that the bottom bed in a CFB boiler is a bubbling bed, but their interpretation of the reasons for this behaviour differs from the above. They found, supported by their literature survey also including the Chalmers boiler, that the reason for the bubbling state was particle segregation, making the bottom-bed particles coarse. Therefore, its transport velocity $u_t$ was higher than the fluidization velocity in the bottom bed, and the bottom bed remains in a bubbling state of fluidization, while the finer fraction circulates. There are many literature references illustrating this segregation. Fig. 4 is an example, showing the size change during an increase of the fluidization velocity in the Chalmers CFB boiler. When the velocity increases, the size of the circulating solids gradually approaches the size of the bottom-bed particles, which is not affected by the velocity change. In this case, like what was pointed out by Wang et al. (2021), the fluidization velocity was below the transport velocity of the bottom bed.

In Fig. 4 the fluidization velocity was below $u_t$, but there is a development towards a circulation of all bed particle sizes as $u_t$ is attained. The circulation flow of particles was not measured, but it is generally known to increase with the fluidization velocity. One could speculate that when $u_t$ is attained the entire bed circulates, and there could not be any bottom bed. However, it can be discussed how relevant the comparison with the transport velocity is. As mentioned above, all bed particles are not exposed to the actual fluidization velocity due to the high through-flow through the bubbles, and only a certain fraction of the particles feels the high gas velocity. Most particles are contained in a kind of “particle phase” in contact with lower gas velocities than those in the bubbles. The only known observation of the structure of a bed at relatively high velocities is that of Werther and Wein (1994) who measured a voidage of about 0.6 in the dense phase between the bubbles at velocities up to 3 m/s in a bed of 0.5x0.5 m cross section, operated at velocities of up to 4 m/s. As pointed out above, the local variation of the gas flow in a high-velocity bubbling bed is important, and the transport velocity, understood as an average over the cross section, is a more representative quantity in connection to a more even, turbulent bed than to a bubbling bed, having a high level of velocity variations.

2.1. A few general comments

To make interpretations easier, most cases mentioned above are operated in devices with vertical walls and primary air only. Com-

![Fig. 3. Images from a cold, two-dimensional bed (dimensions 0.07 x 0.68 m, and the available height is 3.5 m) operated at velocities $u-u_{um}$ from 0.24 to 2.0 m/s with (right) and without (left) tubes. The bed particles were sand, size 0.79 mm, Andersson et al. (1989).](image)

![Fig. 4. Average particle size of samples taken in the bed at 0.65 m height in the Chalmers CFB boiler (upper curves) and in the downcomer from the cyclone (lower curves) at different gas velocities, from Johnsson and Leckner (1995). Two cases, one with 0.44 mm sand particles and one with 0.20 mm sand particles were investigated. (The boiler was operated with coal, but the bed was just established in both cases, so the amount of ashes was small).](image)
mercial CFB boilers normally have tapered walls in the bottom part and are supplied with secondary air. However, often the geometry and the ratio primary/secondary air are chosen to make the fluidization velocity essentially constant in the entire furnace from the bottom to the top. Therefore, the straight-wall case is also representative for industrial applications with tapered walls, although the detailed fluid dynamics differ somewhat.

The Chinese coals, which are used in some investigations, are known to involve problems with size distributions and composition (Li et al. 2008). The extraction of bottom ashes is located where the largest bed particles are found, and there may be a difference between the bed-particle size and the size of the extracted bottom ashes. The bed is not ideally well-stirred. Because of an imperfect control of the input coal-size distribution, in earlier Chinese CFB boilers there is an important tendency to form a pronounced bottom bed. In such units, there could be some bias in the measurement of the bed particle-size distribution because of samples taken from the extracted bottom bed. The idea of researchers from the Tsinghua University (Yue, 2015) was to save auxiliary electric power for fluidization by reducing the amount of bed material. This would predominantly remove part of the coarse particles, forming the bottom bed, without adversely affecting the operation of the boiler and the circulating fine fraction of bed material. This problem depends on insufficient fuel preparation and its significance can be reduced by improving this process.

The impact of the factors mentioned could be substantial, but they do not change the conclusions made by Wang et al. (2021) with respect to the operation of the bottom bed.

3. The transport zone

The vertical pressure distribution from the Chalmers boiler, Fig. 1, with a pressure drop of about 6 kPa is extrapolated to the situation in a conventional large CFB boiler with a furnace height of 43 m and an assumed bed pressure-drop (which is related to the amount of bed material) of 12 kPa by keeping the data in the lower part equal to those in Fig. 1 and extending the height of the transport zone.

The average particle concentration (1-ε) is calculated by,

\[ dp = g(1-\varepsilon)(\rho_s - \rho_g)H \]

For the transport zone, 7000 = 9.81(1-ε)2600*40 gives a particle concentration of (1-ε) = 0.007 (where \( \rho_s \) and \( \rho_g \) are particle and gas densities and \( g \) is gravity).

As pointed out by Leckner (2017), this particle concentration in the transport zone is in the region of pneumatic transport outside of the range of fast fluidization as it is depicted in regime diagrams (for instance, as mentioned above, those of Li and Kwauk 1980; Yerushalmi et al. 1985; Bai et al. 1993). In addition, Bi and Grace (1995) gives the ranges of 0.8 < ε < 0.95 for “fast fluidization”, 0.95 < ε < 0.99 for “core-annular dilute transport” and ε > 0.99 for “homogeneous dilute transport”. In a later publication, Grace et al. (1999), the fast bed region is extended to ~0.8 < ε < ~0.97 in the case of dense suspension upflow. In Fig. 5 the range of operation in the upper part of the furnace of boilers is compared with the regime limit presented by Bi and Fan (1991), described by the limit for accumulative choking, expressed for a particle size of 0.2 mm, inserted in a relationship containing the dimensionless particle size, the Archimedes number.

The diagram shows the transition between “fast fluidization” and “pneumatic transport” (dilute-phase flow). “Accumulative choking” means that back-mixing (because of incipient clustering) of particles becomes important, whereas in (vertical) lean phase pneumatic transport the particles move upwards without significant back-mixing. “Saturation carrying capacity” is the circulation rate in the upper part of a riser related to the concept of accumulative choking. This is in the core of the riser. Back-flow at the walls may occur, but for other reasons. Close to the demarcation curve, on the dilute-phase transport side, Bi and Grace (1995) define a region seemingly agreeing with this behaviour, called “Core-Annular Dilute Phase Flow”. As can be seen in Fig. 5, the operation data from boilers are on the dilute phase transport side of the accumulative choking limit, while the fast fluidization is clearly on the other side of this limit (as it should be).

The picture given in Fig. 5 is not generally accepted. For instance, in a recent publication, authors from the Tsinghua University have evaluated flow regimes in the upper part of a CFB boiler, Cai et al. (2018). They claim “the furnace conditions fall out of the fast fluidization originally defined for the chemical reactors, which is valid for Group A particles at high particle density and high \( G_s \), the fluidization state in the upper furnace of a CFB boiler could still reach to the type A choking state even at low \( G_s \), with the typical features of the strong back-mixing and cluster formation. Namely, the fluidization state can still be regarded as a “fast bed” in a CFB boiler. It is worthy to point out, for the limited studies of the influencing factors mentioned above and the insufficient measurement of \( G_s \) in CFB furnaces, this conclusion also needs future verification”. (\( G_s \) is the circulation flux of particles, kg/m²s). It is difficult to argue against this opinion, but one can agree with the last statement. The reason for the authors’ statement was the even furnace temperature, normally observed in CFB boilers despite heat transfer. However, there are several contributing factors to consider in addition to clustering in the core of the furnace: the wall layers, heat release from combustion, the high heat capacity of the bed material. An evaluation of the impact of all these factors should be carried out to explain the even furnace temperature.

Wang et al. (2021) consider that the fast bed regime occurs in fluid catalytic-cracking reactors but not in CFB boilers because of the different size and density of the particles and other operational differences. Furthermore, they think that CFB boilers do not operate in the transport regime because of the solids reflux that often exists along the height of the furnace above the bottom bed. Instead, they call the mode of fluidization “entrained flow”. This mode is regarded to cover the entire space above the bubbling bed.

This space actually consists of at least two parts: the splash zone and the transport zone with mutually different properties. Table 1 indicates that the splash zone on an average is about dense
enough to behave as fast fluidized according to the fluidization regime diagrams, but the reason for the development of clusters is specific for the flow situation. Here, the particle suspension is produced by the ejection of particles from a bubbling bottom bed, both small and large, caused by the bursting bubbles. The ejected and entrained particles are carried up in the riser aided by the gas jets accompanying the splashing bubbles. When the momentum of a jet dissipates and the velocity decreases, the particles fall back to the bed by themselves because of their weight or promoted by clustering, while smaller particles are entrained upwards by the gas. The splash zone serves as a filter, separating heavier, larger bed particles from finer, circulating particles. This is not a well-defined process, however, and generally valid correlations for the height of the splash zone in the form of a Transport Disengaging Height, introduced by Zenz and Weil (1958) for bubbling beds, has not been found in CFB, Cahyadi et al. (2015). Above the splash zone, the suspension is very dilute (Table 1) and the interaction between particles is limited. Numerical calculation by Nikolopoulos et al. (2021) has identified zones in a furnace where clustering is weak, called “homogeneous” and other zones with higher particle concentration where clustering plays a role, called “heterogeneous”, Fig. 6. The dots inserted in Nikolopoulos’ diagram illustrate the conditions in the upper furnace and in the splash zone, as given in Table 1.

<table>
<thead>
<tr>
<th>Part of bed</th>
<th>Number in Fig. 1</th>
<th>Height H m</th>
<th>Pressure drop, kPa</th>
<th>Particle concentration, 1-e</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bottom bed</td>
<td>1</td>
<td>0.5</td>
<td>4</td>
<td>0.30</td>
</tr>
<tr>
<td>Splash zone</td>
<td>2</td>
<td>2</td>
<td>1</td>
<td>0.02</td>
</tr>
<tr>
<td>Transport zone</td>
<td>3 + 4</td>
<td>40</td>
<td>7</td>
<td>0.007</td>
</tr>
<tr>
<td>Total</td>
<td>———</td>
<td>43</td>
<td>12</td>
<td>———</td>
</tr>
</tbody>
</table>

Fig. 6. The voidage of the upper furnace and the splash zone from Table 1 are shown by round, red markers inserted into the map of Nikolopoulos et al. (2021). The map shows the impact of clusters on the drag. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 7. Impact-probe measurements from the wall layer and in the core of the Turow 235 MW, CFB boiler. The vertical locations for measurements are shown on the boiler profile in the right-hand part of the figure, Johnsson et al. (1999).
There is a difficulty in making generally valid conclusions because the situation is not completely black and white. Rather, it should be regarded from a statistical point of view, accounting for the probability of particle incidents to occur, such as seen in Fig. 7, where measurements with an impact probe in various parts in the furnace of a commercial CFB boiler are shown, indicating the effect of particles.

Fig. 7 shows that the highest activity is found in the splash zone, the lowermost measurement location in the figure. There is very little activity in the upper core, except that particles are transported upwards, but still, now and then, a particle group causes a disturbance on the probe. The activity in the wall layer is always higher than in the core.

The impact measurements distinguish between the upward flow of particles, mostly in the core, and the downward flow, mostly at the wall, except in the upper splash zone, the lower diagrams, where there is a clear flow in both directions. Werther (1993) presents continuous samplings from the Flensburg CFB boiler, illustrating the above statements, giving a time-integrated picture. The reason for the different opinions may be found in the discussion. The reason for the different opinions may be found in the discussion.

The comparison of a few selected observations from researchers on the fluid dynamics of CFB boilers shows a difference in interpretation. The reason for the different opinions may be found in the lack of sufficient information for safe conclusions because these furnaces are large and closed by membrane-tube walls. They are very difficult to access for exploratory measurements.

Fig. 7 shows some examples of measurements through holes made in the fins between the boiler tubes. The project was stopped abruptly when erosion was discovered in the vicinity of these holes.

It can be concluded in agreement with Cai et al. (2018) that further research is required, and relationships explaining the impact of particle size and density, particle concentration, fluidization velocity, and gas properties should be established.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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