Hydrodynamic Properties of High Activated $\gamma$-Al$_2$O$_3$ in a Fixed Bed Reactor

Xuejiao Cao, Ting’an Zhang*, Yan Liu, Yubin Zhang and Weiguang Zhang

Special Metallurgy and Process Engineering Institute, School of Metallurgy, Northeastern University, No.3-11, Wenhua Road, Heping District, Shenyang, Liaoning 110819, China
E-mail:zta2000@163.net

Abstract. The high activated $\gamma$-Al$_2$O$_3$ hydrodynamic properties in the fixed bed are studied in this paper. The factors of gas velocity and the heights of the bed on the axial bed pressure, bed pressure drop and bed voidage are studied. The results indicated that under the same height of fixed bed, the axial pressure decreased along the bed at the same gas velocity. While the pressure drop increased with the gas velocity and the bed height increased. The pressure drop has a good agreement to the Ergun equation. According to the Ergun equation, the bed voidage is regressed at different height of fixed bed. The results revealed that the bed voidage of high-activated $\gamma$-Al$_2$O$_3$ is almost the same in the fixed bed heights of 400 mm, 300mm and 200mm, which is 0.31, 0.32, and 0.32 respectively.

1. Introduction

Primary aluminum is extracted from alumina electrolytic process where alumina and cryolite dissolved and reacted at 950-960 °C[1,2]. In this process, a mass of toxic and noxious flue gas is produced. 8-11kg of hydrogen fluoride (HF) and 4-16 kg of SO$_2$ are emitted correspondingly for 1t aluminum in the flue gas, which has serious hazards on environment and human by way of damaging the lungs and respiratory system[3]. However, new standard of emission standard of pollutants for aluminum industry(GB25465-2010) was issued[4]. The new standard imposes higher requirements. Namely, total fluorine emission decreased from 6 mg·Nm$^{-3}$ to 3 mg·Nm$^{-3}$ and SO$_2$ from 400 mg·Nm$^{-3}$ to 100 mg·Nm$^{-3}$. Thus, it is necessary to remove HF and SO$_2$ further in current aluminum plants.

When considering SO$_2$ removal, almost 90% of flue gas desulfurization in the world is using wet desulfurization technology [5]. But the residual HF in the flue gas will result in equipment corrosion in the desulfurization process and make the subsequent processing more difficulty. Based on this, our research group proposed a new method of using high-activated $\gamma$-Al$_2$O$_3$ for the deep removal on low concentration of HF in a fixed-bed reactor and subsequently a ZnO desulfurization process to remove SO$_2$ [6]. Namely, a new fixed-bed technology is conducted after traditional dry defluorination technology, which can remove completely low concentration of HF and eliminate the effect of HF on subsequent wet desulphurization process.

In this paper, the axial gas pressure, pressure drop, and mixing state of solid particles in fixed bed reactor was systematically studied. Here, changes in gas pressure can reflect gas-solid contact state and the energy dissipation of pumped gas [7]. Moreover, the voidage of solid particles in fixed-bed reactor represents interaction condition of solid-solid and solid-gas. The above two has a significant influence on adsorption defluorination process and reflect the high-activated $\gamma$-Al$_2$O$_3$ hydrodynamic properties, which also has a great significance on engineering amplification design and application of fixed-bed reactor.
2. Methodology

2.1. Principles
Bed Pressure drop in a fixed bed reactor has been researched for many years, and there are many calculate equations, such as Ergun equation \[8\], Lewis equation \[9\], and Kwauk equation \[10\]. The Lewis equation is used for small range of Re (Re<10). The Ergun equation is used for a wider Re range, but it is more complex and needs several parameters \[11\].

\[
\frac{\Delta P}{\Delta L} = f_k \frac{(1-\varepsilon)}{\left(\frac{d_p \varepsilon^2}{\rho_f u^2}\right)}
\]

where \(\Delta P\) is pressure drop across fixed bed, Pa; \(\Delta L\) is the length of fixed bed, m; \(f_k\) is the friction coefficient of fluid in the bed; \(\varepsilon\) is bed voidage; \(d_p\) is the particle diameter, m; \(\rho_f\) is the fluid density, kg·m\(^{-3}\); \(u\) is the superficial fluid velocity, m·s\(^{-1}\); \(\mu\) is the fluid viscosity, N·s·m\(^{-2}\); \(\Re\) is the Reynolds number. The Ergun equation can be used under the condition of \(\Re / (1-\varepsilon) \leq 2,500\).

According to equation (1), (2), (3), the Ergun equation can be written as:

\[
\Delta P = f_k \frac{(1-\varepsilon)}{\left(\frac{d_p \varepsilon^2}{\rho_f u^2}\right)} \Delta L
\]

For some fluid and material, the pressure drop is only the functions of the conditions of the material voidage, the gas velocity and the bed height \[12\]. By using Origin 8.5 software, define the equation in the builder, the bed voidage can be regressed under different velocities.

2.2. Apparatus and Materials
Experiments are conducted utilizing a cold model facility. The facility comprises a fixed bed reactor of 50 mm inner diameter and 500 mm height, an air supply system, and a pressure observation system, as shown in Fig. 1. Four axial pressure observation points were distributed at different height along the reactor: Point 1 is the gas inlet pressure observation point; Point 2, Point 3, Point 4 is 200 mm, 300 mm, and 400 mm from the gas inlet along the axial direction, respectively. The pressure is measured by a 4-20 mA pressure transmitter.

![Figure 1. Experimental apparatus](image-url)
3. Results and Discussion

3.1. Effect of Gas Velocity on the Bed Axial Pressure

Pressure of four axial observation points on the reactor is measured by the pressure transmitter under the conditions of the bed height of 400 mm and the gas velocities of 0.57-1.2 m·s\(^{-1}\). As shown in Fig. 2, pressure observed at the same point increases with the gas velocity increase from 0.57-1.17 m·s\(^{-1}\). When the gas velocity is higher than 1.17 m·s\(^{-1}\), the pressure at the air inlet decrease suddenly, but the pressure in the other positions do not decrease obviously. When the gas velocity is 1.2 m·s\(^{-1}\), the particles in the bed starts moving, the voidage of the bed increases, so the pressure falls down suddenly. Therefore, when the bed height is 400mm, the gas velocity at flood point is 1.2 m·s\(^{-1}\).

![Figure 2. Axial pressure in fixed bed with the bed height of 400mm](image)

3.2. Effect of Gas Velocity on Bed Pressure Drop

The axial bed pressure drop at different parts of the bed is measured under different gas velocities and results are shown in Fig. 3. In the picture, line A is the axial pressure drop between point 1 and 4, line B is axial pressure drop between point 1 and 3, and line C is axial pressure drop between point 1 and 2. As can be seen from Fig. 3, when the bed height is 400 mm, the whole bed pressure drop (Line A) is bigger than the bed inside pressure drop (Line B, and Line C) at the same gas velocity. The bed pressure drop (Line A) increases with the gas velocity increase before the flood point velocity. The bed pressure drop decreases suddenly when the gas velocity reaches the flood point velocity of 1.2 m·s\(^{-1}\).

![Figure 3. Axial pressure drop in fixed bed with the bed height of 400 mm](image)
3.3. Bed Voidage Calculation

According to the Ergun equation (4), the voidage of the axial fixed bed is regressed by the Origin 8.5 software when the fixed bed height is 400 mm. The regression process is executed under the conditions of $d_p = 0.0039 \text{ m}$, $f = 1.293 \text{ kg·m}^{-3}$ and $\varepsilon = 18.28 \times 10^{-6} \text{ N·s·m}^{-2}$, and the results are shown in Figure 4. The results indicate the whole fixed bed voidage is about 0.307, as shown in Fig. 4(a). The bed voidage of different axial parts is also regressed. The results indicate between the gas inlets to the axial distance of 300 mm the bed voidage is 0.311, as shown in Fig. 4(b), while between the gas inlets to the axial distance of 200 mm the bed voidage is 0.314, as shown in Fig. 4(c). The whole bed pressure drop (pressure drop between point 1 and point 4) is recalculated by the Ergun equation with an average voidage of 0.31 in the 400 mm fixed bed. The results show that the relative error is less than 6.04%, given in Table 1.

Table 1. Bed pressure drop in fixed bed reactor with a bed height of 400 mm

| $Q_f$ (m$^3$·h$^{-1}$) | $u_f$ (m·s$^{-1}$) | $\Delta P_{exp}$ (Pa) | $\Delta P_{cal}$ (Pa) | Relative Error (%) |
|----------------------|-------------------|----------------------|----------------------|-------------------|
| 4.0                  | 0.57              | 700                  | 745.02               | -6.04             |
| 5.0                  | 0.71              | 1,100                | 1,093.64             | 0.58              |
| 6.0                  | 0.85              | 1,500                | 1,507.58             | -0.50             |
| 6.5                  | 0.92              | 1,800                | 1,739.04             | 3.51              |
| 7.0                  | 0.99              | 2,000                | 1,986.84             | 0.66              |
| 7.5                  | 1.06              | 2,300                | 2,250.96             | 2.18              |
| 8.0                  | 1.13              | 2,600                | 2,531.41             | 2.71              |
3.4. Effect of Bed Height on the Bed Axial Pressure and Bed Pressure Drop

Pressure of four axial positions on the reactor is measured by the pressure transmitter under different flow velocities with the bed heights of 200 mm and 300 mm. The results are shown in Fig. 5. As can be seen from the Fig. 5 (a), the pressure at axial point 2, 3, 4 is almost the same when the bed height is 200 mm. With the gas velocity increase, the pressure observed at point 2, 3, 4 increases slightly. However, the gas inlets pressure (pressure at point 1) increases with the gas velocity increase from 0.28 m·s⁻¹ to 1.20 m·s⁻¹. While the velocity reaches 1.24 m·s⁻¹, the pressure at point 1 falls suddenly. When the bed height is 300 mm, the pressure obtained at point 3 and 4 are almost the same, because there are no high activated alumina filled in these parts of the fixed bed, so the pressure changes slightly between point 3 and point 4. However, pressure measured at point 1 and point 2 increases with the gas velocity increases from 0.28 m·s⁻¹ to 1.24 m·s⁻¹. When the gas velocity increases to 1.27 m·s⁻¹, the pressure obtained at point 1 decreases suddenly, as shown in Fig. 5 (b). According to the Ergun equation (4), the whole bed voidage of different bed height of 200 mm and 300 mm are obtained, as shown in Figure 6. The results indicate that the bed voidage is 0.319 and 0.324 in the height of 200 mm and 300 mm fixed bed.

![Figure 5. Pressure in different axial parts with different bed heights of 200 mm and 300 mm](image1)

![Figure 6. Bed voidage regression with different bed height of 200 mm and 300 mm](image2)
4. Conclusions
The hydrodynamic properties of high activated $\gamma$-Al$_2$O$_3$ in the fixed bed have been investigated. The axial pressure along the fixed bed decreases along the bed when the gas velocity and height fixed. The bed pressure drop increases with the gas velocity and the heights increases. However, the pressure drop decreases suddenly, when the gas velocity increases to a flood point velocity. The flood point velocities at different bed heights of 200 mm, 300 mm, and 400 mm are 1.20 m·s$^{-1}$, 1.27 m·s$^{-1}$, 1.17 m·s$^{-1}$ respectively. According to the Ergun equation, the bed voidage of high-activated $\gamma$-Al$_2$O$_3$ in different fixed bed heights of 200 mm, 300 mm, and 400 mm is regressed, which is almost the same. The bed voidage is 0.32, 0.32, and 0.31 respectively in the fixed bed heights of 200 mm, 300 mm, and 400 mm.

5. Acknowledgements
The financial support given by the Iron and Steel Joint Fund of National Natural Science Foundation of China Committee (No. U1760120), the National Key R&D Program of China (2017yfc0210403-04), the Fundamental Research Funds for the Central Universities (N182504018), the Fundamental Research Funds for the Central Universities (N2025038) and the Fund of Liaoning S&T Project (20180551008).

6. References
[1] Hou J F, Shi D, Wang Z W, et al. Influence of Additives on bath analysis in aluminum electrolysis. JOM. 2017; 69: 2057-2064.
[2] Cassayre L, Palau P, Chamelot P, Massot L. Properties of low-temperature melting electrolytes for the aluminum electrolysis process: A Review. J. Chem. Eng. Data 2010; 55: 4549-4560.
[3] Stephen CM, Claude L. Community health risk assessment of primary aluminum smelter emissions. Journal of Occupational and Environmental Medicine 2014; 56(58): 33-39.
[4] Emission standard of pollutants for aluminum industry, PRC Standard GB25465-2010
[5] Neveux T, Moulec Y L. Wet industrial flue gas desulfurization unit: model development and validation on industrial data. I & EC Research 2011; 50: 7579-7592.
[6] Zhang, T. A., Liu, Y., Zhang, W. G., et al. China patent, CN 107803108 A, Shenyang (2018).
[7] Baghapour B, Rouhani M, Sharafian A, Kalhori SB, Bahrami M. A pressure drop study for packed bed adsorption thermal energy storage. Applied Thermal Engineering 2018; 138: 731-739.
[8] Ergun S. Fluid flow through packed columns. Chem. Eng. Pro. 1952; 48(2): 89-94.
[9] Lewis W K, Gilliland E R, Bauer W C. Characteristics of fluidized particles. Ind. and Eng. Chem. 1949; 41(6): 1104-1117.
[10] Kwauk M. Generalized fluidization I. Steady-state motion. Scientia Sinica 1963; 12(4): 587-612.
[11] Andrejčíc M, Podgornik A. Effect of pressure drop model implemented for description of pressure drop on chromatographic monolith on estimated adsorbed layer thickness. Chem. Eng. Sci. 2017; 161: 370-381.
[12] Xu S P, Chen Q J, Liang X Y, Ling L C. The hydrodynamic properties of spherical activated carbon in the fixed bed. Carbon 2008; 3: 24-28.