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NUMERICAL SIMULATION ON CARBONATION REACTOR OF CALCIFIED RESIDUE

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Abstract

This paper aimed at studying the carbonation process - the core process of "Calcification-Carbonation", and a venturi carbonation reactor to process calcified residue was innovatively designed and produced. The simulated calculations for dynamic characteristics of gas-liquid-solid three-phase flow were discussed under different superficial gas velocities. Results showed: with the increase of gas velocity, gas holdup increased and solid holdup decreased slightly. Fluid turbulence intensity and collision and coalescence of bubbles were enhanced with the increase of gas velocity, which was beneficial to the mass transfer and reaction among gas, liquid and solid. The measured values agreed well with the simulated calculations obtained by CFD, which indicated that it was feasible to use this model to simulate the gas-liquid-solid three-phase flow field in this reactor and the results of numerical simulation were reliable. The above results provided a theoretical basis for the design of the reactor under a high temperature and a high pressure.

Introduction

Northeastern university puts forward a new method named carbonation process, which treats low-grade bauxite (red mud) and other materials containing aluminium after years of research. That is: firstly, the total Si contained in bauxite and red mud is calcified into Ca-Al-Si compounds (hydrogarnet), then a mixture of calcium silicate, calcium carbonate and aluminum hydroxide is obtained by using CO_2 to process the hydrogarnet, finally the new structural red mud mainly containing calcium silicate and calcium carbonate is obtained by dissolving alunium under low temperature, which can be directly used in the cement industry to achieve the recycle of bauxite resources.

At present, there have been two national invention patents of this technology[1,2]: "Alumina production transformed based on calcification-carbonation method" and "Method for dissolving Bayer process red mud" and Shenyang Aluminum & Magnesium Engineering & Research Institute has evaluated its economic benefits. It indicates that better economic benefits are obtained during the production than other similar ones in China[3-7], especially dealing with the low Al-Si ratio materials (red mud), and it only needs to add the core equipment of carbonation into existing production line, which has a broad prospect.

Despite the advantages above, the two key processes of calcification and dissolution of low grade bauxite and carbonation of calcified residue need to be studied systemically in order to achieve the breakthrough on this new method. In this process, gas-liquid-solid three phases are involved, which is a complicated three-phase system in slurry bed reactor.

In this work, a new Venturi carbonation reactor was proposed and innovative research focused on carbonation process was carried out, which studied mixing effect of gas-liquid-solid three phases and explored change law of flow field in the carbonation reactor of calcified residue. The influence factors and interact rules of three-phase mixing were systemically investigated by using numerical simulation, which provided the theoretical and experimental basis for the further studies.

Mathematical simulation

Mathematical model

In this paper, the interaction among the solid, liquid and gas was simulated by using using Euler three-phase model. Only fluid flow in the reactor was considered and energy transmission was ignored. Mathematical equations are as follows.

Continuity equation:

$$\frac{\partial}{\partial}(\alpha_q \rho_q) + \nabla \cdot (\alpha_q \rho_q \vec{\upsilon}_q) = 0 \quad (1)$$

$q = g, l, s$, Where $\vec{v}_q$ is the velocity of q phase.

Momentum equation of gas phase and liquid phase:

$$\frac{\partial}{\partial t}(\alpha_{g,l}\rho_{g,l}\vec{v}_{g,l}) + \nabla \cdot (\alpha_{g,l}\rho_{g,l}\vec{v}_{g,l}\vec{v}_{g,l}) = -\alpha_{g,l}\nabla p + \nabla \cdot \overline{\overline{\tau}}_{g,l} + \alpha_{g,l}\rho_{g,l}\vec{g} + \sum \vec{F}_{g,l} \quad (2)$$

Momentum equation of solid phase:

$$\frac{\partial}{\partial t}(\alpha_s \rho_s \vec{v}_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s \vec{v}_s) = -\alpha_s \nabla p - \nabla p_s + \nabla \cdot \overline{\overline{\tau}}_s + \alpha_s \rho_s \vec{g} + \sum \vec{F}_s \quad (3)$$

$$\overline{\overline{\tau}}_q = \alpha_q \mu_q (\nabla \vec{v}_q + \nabla \vec{v}_q^T) + \alpha_q (\lambda_q - \frac{2}{3}\mu_q)\nabla \vec{v}_q \overline{\overline{I}} \quad (4)$$

Where $\overline{\overline{\tau}}_q$ is the stress tensor of q phase.

Standard k-ε Mixture Turbulence Model was chosen for simulating the gas-liquid-solid three-phase turbulence.

Geometric model

Venturi jet device is the ZSL（F）hydraulic injector produced by HeBei Sanyang industry FRP Co., Ltd. and the size is shown in fig 1 where throat diameter is 25mm, inlet diameter is 38mm, outlet diameter is 36mm and gas inlet is diameter 20mm. Venturi jet device is connected with the main reactor and the main reactor is made of organic glass, where the inner diameter is 240mm, the effective height is 615mm and the diameter of the overflow port is 100mm.

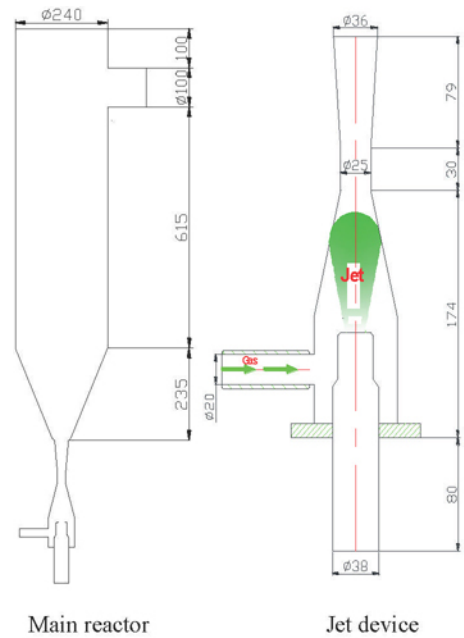


Fig.1 The geometric size of reactor

Physical parameters

In this paper, the fluid was regarded as incompressible fluid and energy equation was ignored, i.e. heat transfer was not considered. Wall condition was no-slip and heat insulating. Ambient atmospheric pressure was 101325Pa and gravity acceleration was 9.81m/s^2. Other physical conditions are shown in Table 1.

Table 1 Relative physical parameters

material	density kg/m^3	viscosity Pas	particle diameter μm
air	1.225	1.7894×10^{-5}	
water	1000	1.003×10^{-3}	
glass bead	2477		176

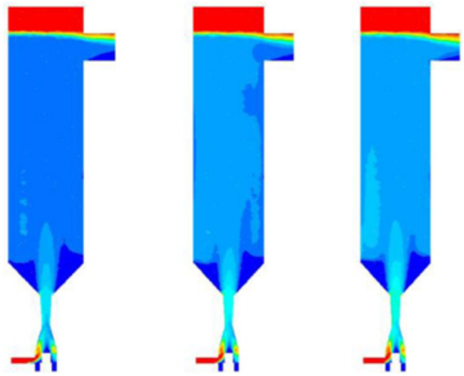
Boundary condition

Velocity-inlet boundary condition was used for gas and liquid inlet, fully developed outflow boundary condition was used for fluid outlet and no-slip boundary condition was used for wall.

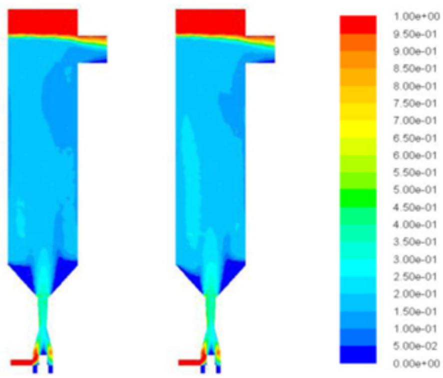
Results and discussion

Influence of superficial gas velocity on gas holdup

Experimental conditions are as follows: in the gas-liquid-solid three-phase system, the superficial liquid velocity is 5.47 m·s^{-1}, superficial gas velocities are 2.63 m·s^{-1}, 3.54 m·s^{-1}, 4.42 m·s^{-1}, 5.31m·s^{-1}, 6.19 m·s-1 and liquid-solid ratio is 10:1.



(a)U_G=2.63 m·s^{-1} (b)U_G =3.54 m·s^{-1} (c)U_G =4.42 m·s^{-1}



(d)U_G =5.31 m·s^{-1} (e)U_G =6.19 m·s^{-1}

Fig.3.1 Gas contours with different superficial gas velocity of x=0 plane

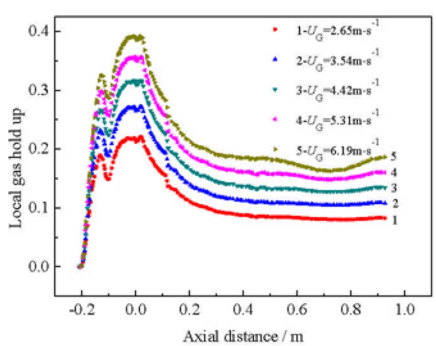
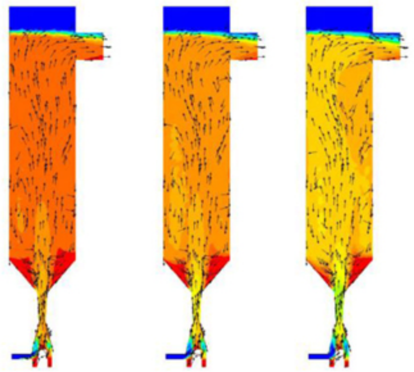


Fig.3.2 Local gas holdup under different superficial gas velocity

Figure 3.1 (a~e) shows the gas phase distribution under the conditions of different superficial gas velocities at the x=0 plane and Figure 3.2 shows the corresponding local gas holdup. As can be seen from the figures, gas holdup in the carbonation reactor of calcified residue monotonically increases with the increase of gas velocity. This is mainly because under the same gas intake area, intake flowrate increases with the increase of gas intake velocity, and then the number of bubbles increases, which causes gas holdup increases at the same plane. Besides, gas holdup presents the trend of uniform distribution. Thus the Venturi jet device in this study has a good effect in the bubble disintegration and dispersion, which provides the theoretical and experimental basis for the further design of high pressure and temperature reactor. However, there exists a suitable range for the increase of gas holdup. Related research showed that gas holdup in the fluidized bed could not be more than 0.3, otherwise gas and liquid would merge with each other and the dispersive gas-liquid two-phase flow, in which liquid was the continuous phase, would be destroyed[8]. As can be seen from figure 3.2, the average gas holdup in the carbonation reactor of calcified residue is lower than 0.3 and in this study, the maximum of average gas holdup is 0.21. Therefore the gas-liquid two-phase flow using the liquid phase as the continuous phase in the reactor can still be stable, which ensures that the solid can achieve continuous motion together with the liquid phase.

Influence of superficial gas velocity on velocity field

Experimental conditions are as follows: in the gas-liquid-solid three-phase system, the superficial liquid velocity is 5.47 m·s^{-1}, superficial gas velocities are 1.77 m·s^{-1}, 3.54 m·s^{-1}, 6.19 m·s^{-1} and liquid-solid ratio is 10:1.



U_G=1.77 m·s^{-1} U_G=3.54 m·s^{-1} U_G=6.19 m·s^{-1}

Fig.3.3 Vectors of liquid velocity

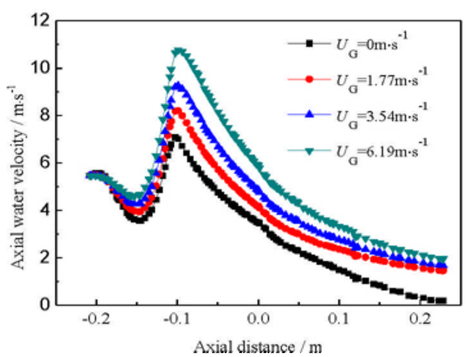


Fig.3.4 Axial liquid velocity at different U_G

Figure 3.3 shows the liquid velocity vector maps under different superficial gas velocities and the background images are the corresponding liquid phase distribution maps. It can be drawn from the figure that the fluid in the reactor presents circulation flow, which flows upward in the center of bed and downward near the wall. This is mainly because of the gravity sedimentation.

Figure 3.4 shows the axial velocities of liquid on the central axis. The abscissa is the axial position of carbonation reactor of calcified residue and the ordinate is the vertical velocity of liquid phase. It can be concluded from the figure that liquid velocity increases with the increase of superficial gas velocity, which is because with the increase of superficial gas velocity, the energy in the system increases and then the turbulence intensity is enhanced. It can be also drawn that the peak velocity appears in the throat of the Venturi jet device, which is determined by the minimum throat diameter.

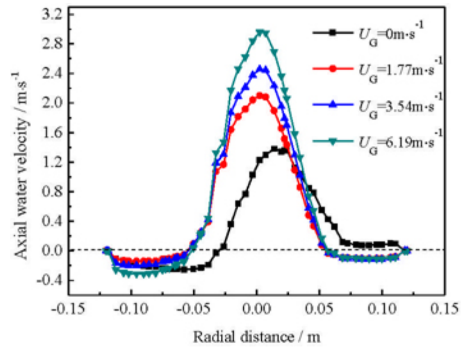


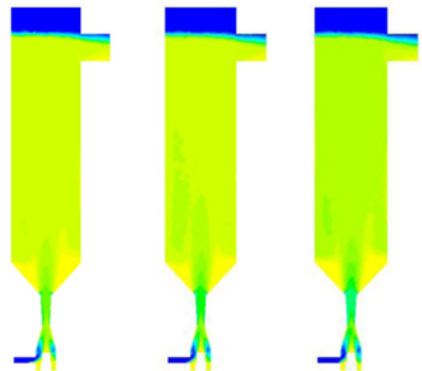
Fig.3.5 Radial liquid velocity at different U_G

Figure 3.5 shows the radial velocities of liquid and gas at the interface between the cone and the cylindrical main reactor. The abscissa is the radial position of carbonation reactor of calcified residue and the ordinate is the vertical velocity of liquid phase. As can be seen from the figure, the liquid velocity increases with the increase of gas velocity. Due to the jet, the liquid velocity reaches the peak near the central axis, decreases along both sides and becomes negative near the wall. This is mainly because under the influence of gravity, fluid in the reactor presents circulation flow as shown in the figure 3.3, which flows upward in the center of bed and downward near the wall. This is also a specific characteristic of the new reactor, which is the circulating fluidized bed between traditional fluidized bed and transport bed. It can be also drawn from the figure that the peak of liquid velocity is off-center without gas while the peak appears in the center after gas passes into liquid, which proves that gas phase is the main turbulence phase in the reactor.

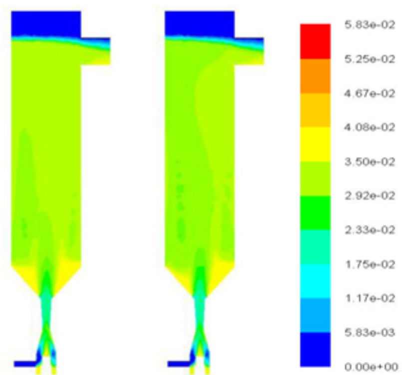
In conclusion, when gas velocity increases, fluid turbulence intensity in this reactor is enhanced and the increased bubbles make the effect of collision and coalescence enhanced, which is beneficial to the mass transfer and reaction among gas, liquid and solid.

<u>Influence of superficial gas velocity on solid holdup</u>

Experimental conditions are as follows: in the gas-liquid-solid three-phase system, the superficial liquid velocity is 5.47 m·s^{-1}, superficial gas velocities are 2.63 m·s^{-1}, 3.54 m·s^{-1}, 4.42 m·s^{-1}, 5.31m·s^{-1}, 6.19 m·s^{-1} and liquid-solid ratio is 10:1.



(a)U_G=2.63 m·s^{-1} (b)U_G =3.54 m·s^{-1} (c)U_G =4.42 m·s^{-1}



(d)U_G =5.31 m·s^{-1} (e)U_G =6.19 m·s^{-1}

Fig.3.6 Solid contours with different superficial air velocity of x=0 plane

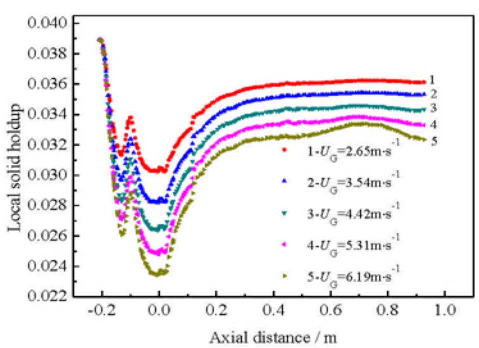


Fig.3.7 Local solid holdup under different superficial air velocity

Figure 3.6(a~e) shows the solid phase distribution under the conditions of different superficial gas velocities at the x=0 plane and Figure 3.7 shows the corresponding local solid holdup. It can be drawn from the figure that solid holdup decreases slightly with the increase of superficial gas velocity, which is consistent with the literature [9] but inconsistent with the literature [10]. This is mainly because with the increase of superficial gas velocity, the effect of bubble coalescence and turbulence is enhanced so that the liquid velocity and the number of entrained particles increase, which causes the slight decrease of solid holdup. It can be also seen from the figure that the particle concentration increases at the bottom of the cone, which is mainly because of the gravity sedimentation. Solid holdup presents the trend of uniform distribution in the carbonation reactor of calcified residue. Therefore, the Venturi jet device has a good effect in mixing of gas-liquid-solid three phases and the results will provide the theoretical basis for the further design of high pressure and temperature reactor.

Comparison between the measured values and simulated calculations of average gas holdup

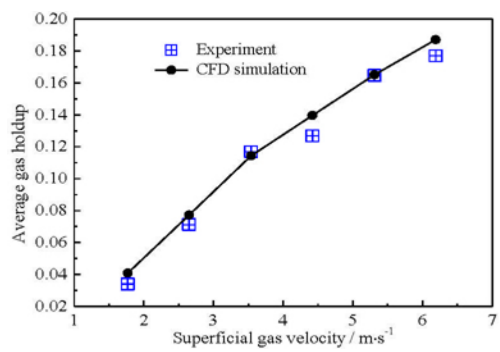


Fig.3.8 The average of gas holdup compare between experiment and CFD simulation under different superficial gas velocity

Figure 3.8 shows the comparison between the measured values obtained by pressure difference method and the simulated calculations obtained by CFD. It can be drawn from the figure that simulated calculations agree well with the measured values and the average gas holdup increases with the increase of superficial velocity. Although few measured values are a little different from the simulated calculations, they present the same trend and the error is within the allowable range. So it is feasible to simulate the influence of superficial gas velocity on average gas holdup and the gas-liquid-solid three-phase flow field in the carbonation reactor of calcified residue by using

Fluent software and Eulerian three-phase flow model.

Conclusion

(1) Gas holdup increased with the increase of superficial gas velocity. However, related research showed that gas holdup in the fluidized bed could not be more than 0.3. In this study, the maximum of average gas holdup is 0.21, so the gas-liquid two-phase flow using the liquid phase as the continuous phase in the reactor can still be stable, which ensures that the solid can achieve continuous motion together with the liquid phase.

(2) With the increase of gas velocity, fluid turbulence intensity in this reactor is enhanced and the increased bubbles make the effect of collision and coalescence enhanced, which is beneficial to the mass transfer and reaction among gas, liquid and solid.

(3) Solid holdup decreased slightly with the increase of superficial gas velocity and presented the trend of uniform distribution in the carbonation reactor of calcified residue. It is indicated that the Venturi jet device has a good effect in mixing of gas-liquid-solid three phases.

(4) The measured values obtained by pressure difference method agreed well with the simulated calculations obtained by CFD. It is indicated that it is feasible to use this model to simulate the gas-liquid-solid three-phase flow field in the carbonation reactor of calcified residue and the results of numerical simulation are reliable.

In conclusion, the new Venturi carbonation reactor in this study has a good effect in mixing of gas-liquid-solid three phases and the bubble disintegration and dispersion. The results will provide the theoretical and experimental basis for the further design of high pressure and temperature reactor.

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