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Experimental study on the Gas Bypassing Fraction in the Production of Polycrystalline Silicon Granules

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The gas bypassing fraction (γ_{RD} and γ_{DR}) is an important parameter in an internally circulating fluidized bed. In this work, the gas bypassing fraction was experimentally studied by gas tracing and material balance method. The effect of structure of gas distributor, superficial gas velocity (U_R and U_D), diameter of orifices (d_{or}) on the side wall of the riser and height of the riser (H_R) on the gas bypassing fraction were studied in an internally circulating fluidized bed reactor. Compared with tubular distributor, the flat plate distributor can provide less gas bypassing fraction and reduced the solid forming on the heating surface. When U_R and U_D increased, γ_{DR} increased and γ_{RD} decreased. γ_{DR} increased obviously and γ_{RD} has small variation with increasing of d_{or}. When H_R increased, γ_{DR} first increased and then decreased while γ_{RD} first decreased and then increased. γ_{DR} had a maximum with the riser height at 265mm when the riser height varied in the range of 235mm to 295mm. The optimized structure and operating condition of ICFB was proposed in this study.

1. Introduction

High purity polycrystalline silicon is main raw material for making solar cell (Surek, 2005). Silicon beads are produced by chemical vapor deposition (CVD) of trichlorosilane and hydrogen on the seed particles in a CVD reactor. Siemens process is the most dominating method for high purity polycrystalline silicon production (Li and Zhang, 2010). As a mature technology, it has many advantages such as high purity of the product and easy to control, but the high energy cost and the batch operation lead to the low production efficiency. The internally circulating fluidized bed (ICFB) was proposed to improve the efficiency of the process. A draft tube has been used as riser to divide the particle circulation within the ICFB. Particles flow upwards in the riser and flow downwards in the downcomer. The effective contact area of gas and solids could be increased remarkably. And longer residence time of fine particles in the annulus section may improve the heat transfer (Riley and Judd, 1987) compared with Siemens process. But the reaction gas forms a bypassing into the downcomer. Because the

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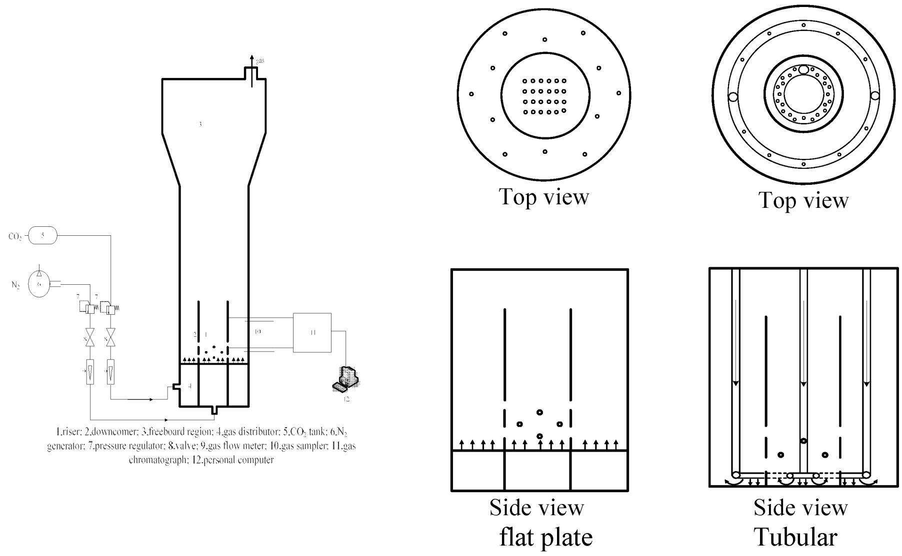
temperature in the downcomer is higher than 900℃, the reaction could occur and form solid of silicon on the heating surface. It could perform negative effect on heat transmission in operation of the ICFB (Kinoshita et al., 1987).

The gas bypassing fractions (γ_{RD} and γ_{DR}) where the subscripts RD and DR denoted the gas bypassing from the riser to downcomer and that gas from downcomer to riser are the important parameters, which depends on operating condition and structure of the ICFB (Yang and Keairns, 1982). The experimental results reported by Song and Kim (1997) showed that the conical tuyere distributor is the most suitable one for a circulating fluidized bed in the used three different types of gas distributor. Ahn et al. (1999) found that the orifice diameter greatly affects the gas bypassing fraction from the annulus section to the draft tube. Shih and Chu (2003) found that the gas bypassing fractions depended primarily on the solids circulation rate (G_s), superficial gas velocity in the riser. Because of the high temperature in the reaction area, the reactor should be made of quartz which with high temperature resistance and has stable properties. The problem of the reactor sealing could not be solved by using the traditional plate distributors which were connected with the apparatus by flanges. The aim of this work is to experimentally determine the effect of the structure of distributor, the diameter of orifices on the draft tube, the height of draft tube and operating condition on gas bypassing fraction in ICFB reactor. A new type of tubular distributor was innovated in this work, the gas bypassing fraction was also experimentally tested, and compared with other distributors used in this work.

2. Experimental

2.1 Apparatus

Experiments were carried out in a Plexiglas ICFB reactor (120 mm I.D. and 800 mm high) as shown in Fig.1 (a). A column riser (70 mm I.D.) with 24 orifices of 10 mm diameter on the side wall was fixed on the bottom of the ICFB. For supplying air into the reactor, two kinds of gas distributor were used in the experiment as showed in Fig.1 (b). The flat plate gas distributor had 24 orifices of 0.8 mm diameter in the riser area and 12 orifices of 0.8 mm diameter in the downcomer area. The tubular distributor with 24 orifices of 0.8 mm diameter in the riser area and 12 orifices of 0.8 mm diameter in the downcomer area were directly connected the compressor. An expanded section (250 mm I.D. and 400 mm height) was installed in the top of the reactor to reduce particle entrainment. Silica beads (d_p=300 μm, ρ_p=2.71 kg/m^3, U_{mf}=0.051m/s), as model particles were used in the experiment. U_{mf} is the minimum fluidization velocity and it depends on the physical property of particles in ICFB. N_2 and CO_2, as the tracer gas which took the place of trichlorosilane and hydrogen, were fed continuously into riser and downcomer, separately. To obtained the gas bypassing fraction, gas samples were taken at the outlets of the riser and downcomer section by sampling probe. The concentrations of the samples were analyzed by a gas chromatograph (GC7890, TDX-01). Several experimental conditions, such as gas velocity in the riser area (U_R) and in the downcomer area (U_D), heights of riser (H_R) and diameters of orifices (d_{or}) were tested in this study.



a. Structure of the ICFB   b. Gas distributors

Fig.1. Scheme of the experimental apparatus

2.2 Measuring method

Gas bypassing fractions (γ_{DR} and γ_{RD}) were measured based on the method proposed by Song and Kim (1997). The gas bypassing fraction γ was defined as the ratio of the inlet gas flow rate and bypassing gas flow rate:

$$\gamma_{RD} = \left(Q_{RD} / Q_{R1}\right) \times 100\% \tag{1}$$

$$\gamma_{DR} = \left(Q_{DR} / Q_{D1}\right) \times 100\% \tag{2}$$

Q_{R1} and Q_{D1} were the inlet flow rate of the riser and downcomer, respectively. With the assumption that tracer concentration of the bypassed gas is equal to that of the corresponding inlet gas ($x_{R1}=x_{RD}=1$, $x_{D1}=x_{DR}=0$), the unknown flow rate (Q_{R2}, Q_{D2}, Q_{RD}, Q_{DR}) can be calculated from mass balances as shown in Fig.2. Q_{R2} and Q_{D2} can be calculated from mass balances and N_2 mass balances of the two regions:

$$Q_{R2} = \frac{x_{D2}\left(Q_{R1} + Q_{D1}\right) - Q_{R1}}{x_{D2} - x_{R2}} \tag{3}$$

$$Q_{D2} = \frac{Q_{R1} - x_{R2}\left(Q_{R1} + Q_{D1}\right)}{x_{D2} - x_{R2}} \tag{4}$$

Q_{RD} and Q_{DR} can be calculated from the mass balances and N_2 mass balances of the riser：

$$Q_{RD} = Q_{R1} - x_{R2}Q_{R2} \tag{5}$$

$$Q_{DR} = (1 - x_{R2})Q_{R2} \tag{6}$$

γ_{DR} and γ_{RD} can be obtained from equation (1) and equation (2).

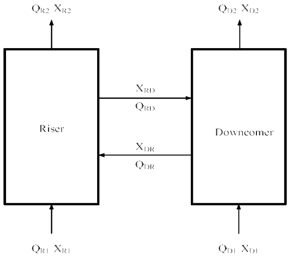


Fig.2. Mass balance of gas bypassing

3. Results and discussion

3.1 Effect of gas distributor and gas velocity

The effect of U_R on γ_{RD} and γ_{DR} with two kinds of distributor was given in Fig.3. The fluidization number was used to describe the changes of the inlet gas velocity. The fluidizations number, U_R/U_{mf} increased with the increase of the inlet gas velocity. γ_{DR} increased with the increase of U_R/U_{mf} for both kinds of gas distributor and γ_{RD} decreased with the increase of U_R/U_{mf}. As reported by Song and Kim (1997) and Shih and Chu (2003), γ_{DR} depends on the pressure drop across the orifices on the side wall of the riser. When U_R increased, the solid holdup in the riser decreased rapidly which resulted in a decrease of fluid density in the riser and resulted in an increase of the pressure drop between the riser and downcomer. Thus, more particles and gas flowed from the downcomer to the riser through the orifices. As observed in Fig.3, γ_{RD} and γ_{DR} with a tubular distributor were larger than that with a flat plate distributor. When tubular distributor was used, gas was introduced through the tubular to the bottom of the bed as shown in Fig.1 (b). The gas bounced back and flowed upwards. The diameter of jet gas flow was larger compared with flat plate distributor. As a result, γ_{RD} increased and γ_{RD} decreased with an increase in U_R. And γ_{RD} and γ_{DR} with a flat plate distributor were lower than that with a tubular distributor.

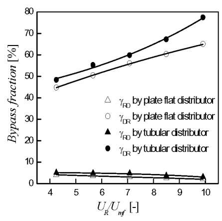


Fig. 3 Effect of U_R/U_{mf} on γ_{RD} and γ_{DR}

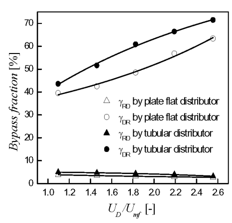


Fig. 4 Effect of U_D/U_{mf} on γ_{RD} and γ_{DR}

The measured results of the effect of U_D on gas bypassing fractions (γ_{RD} and γ_{DR}) for both kinds of gas distributor are shown in Fig.4. The bypassing fraction γ_{DR} increased and γ_{RD} decreased with the increase of the fluidization number U_D/U_{mf} for both kinds of gas distributors. The reason is that when U_D increased, the circulation rate of the particle in the fluidized bed increased. The large circulation rate will increase the part of the inlet gas in the downcomer area flowed into the riser through the orifice on the side wall of the draft tube, which results in an increase of γ_{DR} as reported by Shih and Chu (2003). The experimental results also showed that the gas bypassing fractions, γ_{RD} and γ_{DR} with the tubular distributor were higher than that with flat plate distributor.

3.2 Effect of the orifice diameter on the side wall of the draft tube

The experimental results of d_{or} on γ_{RD} and γ_{DR} were given in Fig.5. When d_{or} increased from 8 to 16 mm, γ_{DR} increased and γ_{RD} kept almost stable. With an increasing d_{or}, it becomes easier for the gas streams and particles to pass through orifices bidirectionally due to resistance around the orifices reduced. Therefore, γ_{DR} and the solid holdup in the downcomer increased. The solid circulation rate increased remarkably because of higher density difference between riser and downcomer and γ_{RD} reached equilibrium in this point. The exorbitant solid holdup in the downcomer, however, could weak the radial mixing behaviour which has positive effect on heat transfer of particles gradually. Milne et al. (1992) have reported that there was little effect of orifice diameter on the gas bypassing. That was due to the opening ratio in this study was 3 times higher than that used in their work. The phenomenon of solids cluster near the orifices was not obvious in this study.

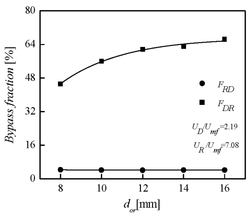


Fig.5 Effect of d_{or} on γ_{RD} and γ_{DR}

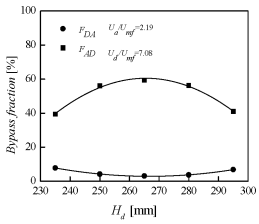


Fig.6 Effect of H_R on γ_{RD} and γ_{DR}

3.3 Effect of height of the draft tube

The effect of H_R on γ_{RD} and γ_{DR} was shown in Fig.6. γ_{DR} first increased and then decreased with increasing H_R from 235 mm to 295 mm. The trend of γ_{RD} was opposite to γ_{DR}. As experimentally observation, the bed height in downcomer increased when H_R increased. It will increase the resistance of the gas flow in downcomer section. As a result, γ_{DR} increased and γ_{RD} decreased when H_R increased from 235 mm to 265 mm. When H_R exceeded 270 mm, G_s decreased rapidly due to the gas flow rate in riser was limited. The gas bypassing was closely related to the particle circulation rate through the orifices. As a result, the γ_{DR} decreased and γ_{RD} increased when H_R increased from 265 mm to 290 mm.

4. Conclusion

The effect of operating condition and structure on γ_{RD} and γ_{DR} has been experimentally determined in this study. The main conclusions are as follows.

a) γ_{DR} increased and γ_{RD} decreased with an increase in U_R and U_D.

b) γ_{DR} and γ_{RD} with a flat plate distributor were smaller than that with a tubular distributor.

c) γ_{DR} increased obviously with increasing d_{or} and γ_{RD} kept almost stable around 4.39%.

d) γ_{DR} first increased and then decreased with increasing H_R, γ_{RD} was opposite to γ_{DR}.

According to the hot state experimental studies, solid of silicon formed on the heating surface when γ_{RD} exceeded 10%. The value of γ_{RD} was controlled below than 8.0% in this study which can fulfill the requirement for polycrystalline silicon production.

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