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Hydrodynamic behaviors of an internally circulating fluidized bed with wide-size-distribution particles for preparing polysilicon granules

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ABSTRACT

An internally circulating fluidized bed (ICFB) reactor was proposed for preparing polysilicon granules to prevent silicon deposition on the heating wall. The fluidization behaviors of the ICFB with wide-size-distribution (WSD) particles were studied, with focus on the gas bypassing and solid circulation rate between the riser and downer. The results showed that the segregation of WSD particles was effectively inhibited by the circulation of particles, and the gas bypassing fraction from the riser to downer, γ_{RD}, was lower than 4%. Using these experimental results, the concentration of $SiHCl_3$ in the reaction zone was estimated to be 7.3–111.0 times larger than that in the heating zone, thus the silicon deposition on heating walls could be effectively suppressed. At normal operating conditions, the solid circulation rate G_s was larger than the minimum value $G_{s,min}$ required by heat supply, thus the ICFB is very promising to solve the heat supply problem without significant silicon deposition on the heating walls in a fluidized bed reactor.

1. Introduction

High purity polysilicon is the main material for the photovoltaic (PV) industry. In the past decade, the global production of solar cells has greatly increased and many efforts have been devoted to preparing high purity polysilicon. Nowadays about 80% of the worldwide polysilicon is produced by the modified Siemens process, which is a batch operation and has a low production and high energy consumption. Therefore many efforts have been made to develop new processes for the polysilicon production. Among these processes, the process using a fluidized bed reactor (FBR) is a very promising alternative to the Siemens process [1]. It has been demonstrated that the direct power consumption of the FBR process using $SiHCl_3$ as reactant is less than half of that of the Siemens process [2]. The production of polysilicon in an FBR greatly increases as the effective deposition area of particles in an FBR is 2–3 orders of magnitude higher than that of the silicon rods in a Siemens bell jar reactor. In addition, our previous studies showed that zero net by-production of $SiCl_4$ can be realized in an FBR, thus the production process can be significantly simplified [3,4].

Although the FBR process has many advantages for the polysilicon production, it suffers from silicon deposition on the reactor wall. The chemical vapor deposition (CVD) of $SiHCl_3$ to silicon is very sensitive to the reaction temperature, therefore the silicon deposition rate is faster on the heating wall if its temperature is higher than that of particles in the bed. The silicon deposition on the heating walls will severely affect the heat transfer and the stable operation of the reactor. To solve this problem, a new structure of internally circulating fluidized bed (ICFB) with separate heating and reaction zones divided by a centrally located draft tube was proposed [5,6]. The ICFB has many advantages such as reduced height and construction cost, smaller heat loss and longer residence time of particles, and therefore has been widely utilized in coal combustion [7,8], coal gasification [9], incineration of solid wastes [10,11] and desulfurization [12,13]. When the ICFB is used for polysilicon production, the reactant gas ($SiHCl_3$ and H_2) is pumped into the reaction zone (draft tube) and the fluidizing gas (H_2) is pumped into the heating zone (annulus), thus the wall deposition can be significantly reduced due to the absence of $SiHCl_3$ in the heating zone.

When the ICFB is used for polysilicon production, the following three key issues must be investigated: (1) The fluidization behaviors of large particles with wide-size-distribution, which are formed by a continuous deposition of silicon, need to be studied, and a good quality of fluidization is required to avoid dead bed or particle segregation; (2) the gas bypassing from the reaction zone to the heating zone occurs due to pressure fluctuations and must be quantitatively studied to ensure that $SiHCl_3$ in the heating zone is low enough to avoid silicon deposition on the heating wall; and (3) the solid circulation rate between the heating zone and reaction

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zone needs to be studied to assure that enough heat is transferred by hot particles from the heating zone to the reaction zone. These issues were investigated in the present work in a cold-mode ICFB with WDS particles, and the segregation behavior, solid holdup, gas bypassing and solid circulation rate were measured under different operating conditions.

2. Experiments

2.1. Apparatus

The cold-model ICFB reactor (300 mm i.d. × 2500 mm height) was made of plexiglass, and its schematic is shown in Fig. 1. By adding a centrally located draft tube (150 mm i.d. × 750 mm height), the reactor was divided into two zones, namely a central zone as a riser and an annulus as a downer. The particles moved downwards in the downer, then circulated to the riser through the orifices (30 holes × 25 mm i.d.) on the lower part of the draft tube under the driving force due to the different solid holdup in the two zones. In an ICFB reactor for polysilicon production, the riser is used as the reaction zone and the downer is used as the heating zone; and the mixture of $SiHCl_3$ and H_2 is pumped into the reaction zone and only H_2 is pumped into the heating zone.

Glass beads were used as fluidized particles because they have similar density to that of silicon granules. Two types of particles were compared, namely the narrow-size-distribution (NSD) particles of 0.2–1.0 mm (packing density: 1567 kg/m^3, U_{mf} = 0.128 m/s), and WSD particles of 0.45–2.5 mm (packing density: 1585 kg/m^3, U_{mf} = 0.484 m/s). The size distributions of the particles are shown in Fig. 2.

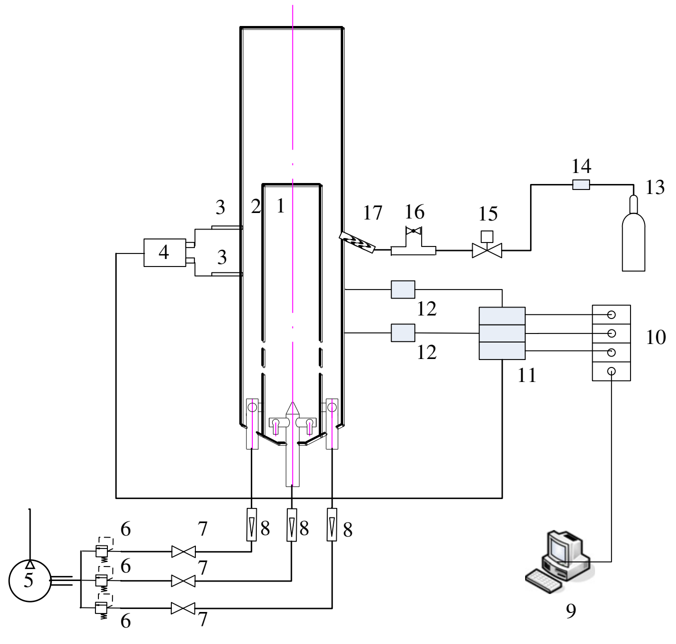

Fig. 1. Schematic of the experimental apparatus. 1 — draft tube; 2 — annulus; 3 — pressure tap; 4 — pressure sensor; 5 — compressor; 6 — pressure regulation; 7 — value; 8 — gas flow meter; 9 — PC; 10 — A/D board; 11 — terminal board; 12 — infrared thermometer; 13 — cylinder; 14 — pressure reducer; 15 — solenoid value; 16 — air escape value; 17 — solid hopper.

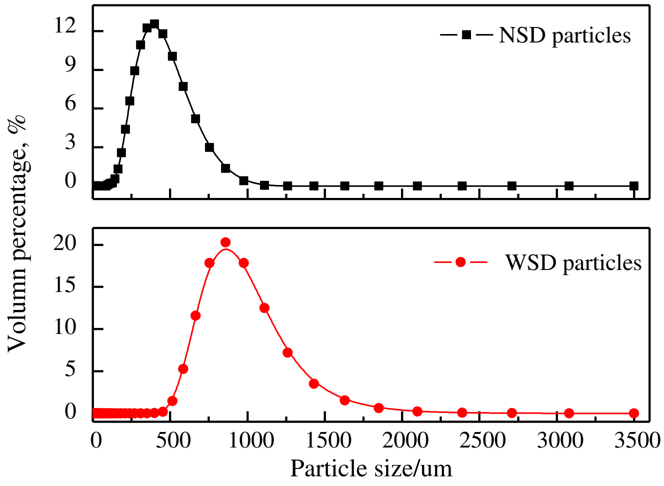

Fig. 2. Size distributions of NSD and WSD particles.

2.2. Measuring method

2.2.1. Solid circulation rate

The solid circulation rate G_s was important to describe the heat transfer capability between the heating and reaction zones. The value of G_s was determined by the solid holdup in the downer ε_D and average particle velocity u_p by

$$G_s = \rho_p \varepsilon_D u_p \tag{1}$$

where ρ_p was the density of particles.

The solid holdup in the downer ε_D was measured by the differential pressure method using two pressure transducers fixed on the reactor wall, as shown in Fig. 1. The relationship between the pressure drop ΔP_b and ε_D was:

$$\Delta P_b = \left[\rho_p \varepsilon_D + \rho_f (1 - \varepsilon_D)\right] gH \tag{2}$$

where ρ_f was the gas density, and H was the distance between two transducers. As $\rho_p \gg \rho_f$, Eq. (2) was simplified to:

$$\varepsilon_D = \Delta P_b / \left(\rho_p gH\right). \tag{3}$$

The average particle velocity u_p in the downer was determined by a thermal tracing method. Hot particles of about 160 °C were injected into the downer with compressed air through a solenoid valve, and the change in the bed temperature was measured by two infrared temperature probes, as shown in Fig. 3. Then u_p was calculated by:

$$u_p = \Delta h / \Delta t \tag{4}$$

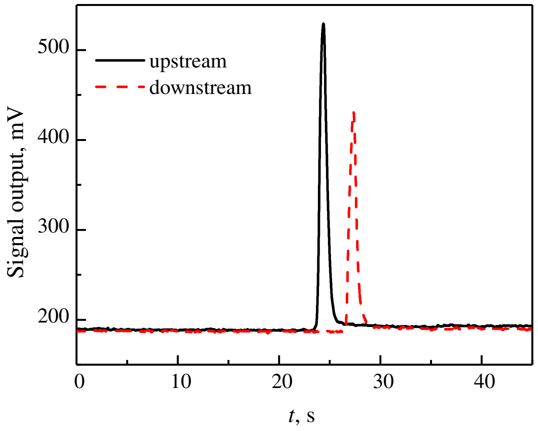

Fig. 3. Typical signals of the infrared temperature probes (U_D / U_{mf} = 1.75, U_R / U_{mf} = 0.65).