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Cold-modeling study of a circulating fluidized-bed reactor for flue gas desulfurization (FGD)

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Abstract: Short residence time of the sorbent in the gas stream and formation of a dense layer of reaction product surrounding its surface influence the sulfur removal efficiency. A practical means of improving the process performance is to employ fluidized-bed reaction in replacement of entrained-bed reaction on normally used in cool-side desulfurizaiton. This paper describes cold-modeling study of a circulating fluidized-bed reactor. Several aspects of the problem are discussed: fluidization behavior of CaO, attrition of the sorbent and solids entrainment from the fluidized bed. Mechanisms and key controlling parameters are identified, and an integral model based on rate of attrition and mass balance is developed for predicting steady-state mass flows and particle size distributions of the system. A process flow scheme is finally presented for conducting desulfurization tests in the second stage of the study.

Key words: circulating fluidized-bed reactor; flue gas desulfurization; cold-modeling study

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Introduction

In the coolside sorbent injection desulfurization, powdered lime and water spray are injected into the flue gas before it enters the particulate collecting equipment. Compared to conventional wet-scrubbing, coolside sorbent injection desulfurization is simpler in flow scheme and less in capital cost. It is especially suitable for cases where FGD installation space and remaining boiler life are limited.

Coolside sorbent injection desulfurization is normally carried out in the form of entrained-bed reaction. The contact time between the two phases is short(~2 sec.), and surface of the lime particles is easily covered by a dense layer of reaction product which hinders the diffusion of SO_2 and O_2 to the inner CaO core. As a consequence, sulfur capture is usually only about 50% and Ca utilization about 25% which influence the process economics.

The employment of a circulating fluidized-bed reactor in replacement of an entrained-bed one can greatly increase the retention time of the solid phase in the system. At the same time, the attrition of lime particles in the fluidized-bed reactor system helps remove the reaction product from the sorbent surface. High sulfur capture and Ca utilization can therefore be expected.

Powdered lime is an adhesive material with properties different from limestone, sand or coal. In developing circulating fluidized-bed technology, it seems necessary to conduct cold-modeling test first to provide a sound technical basis for the desulfurization tests to be performed later.

1 Cold-modeling apparatus

1.1 Flow scheme (Fig.1)

Air enters the reactor and fluidizes the lime bed. Gas leaving the reactor enters the 1st cyclone where most of the entrained lime is removed. The gas then enters the 2nd cyclone for the final removal of particulates. The gas is then washed and vented to the atmosphere through a vacuum pump.

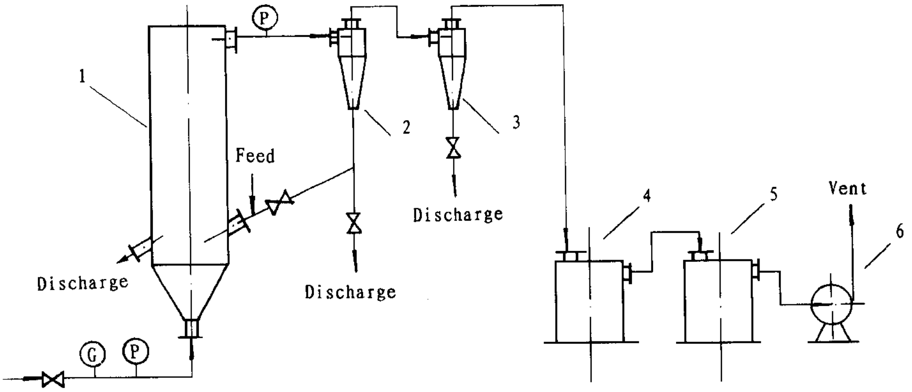

Fig. 1 Flow scheme

1. reactor; 2. 1st cyclone; 3. 2nd cyclone; 4. wash bottle; 5. buffer bottle; 6. vacuum pump

Most of the lime collected in the 1st cyclone is recycled to the reactor, while the remaining part as well as all of the lime collected in the 2nd cyclone are discharged away.

Fresh lime is fed into the system at the leg of the 1st cyclone. Bed lime can be discharged from the reactor through a port located at the lower part of the reactor wall.

1.2 Reactor

Reactor is made of polymethylmethacrylate tube of I.D. 60 mm and height 1220 mm with a perforated plate as gas distributor.

Table 1 Physical behavior and flow characteristics of powdered lime

Specific surface area	1.33 m^2/g
True density	2.37 g/cm^3
Loose bulk density	0.805 g/cm^3
Tapped bulk density	1.089 g/cm^3
Angle of repose	53.3
Angle of slide	42.3
Internal coefficient of friction under vertical load 1000g	
Normal compressive force = 255.27×10^{-3} N/cm^2	
Shear strength = 240.32×10^{-3} N/cm^2	
External coefficient of friction under vertical load 1000g	
Normal compressive force = 207.31×10^{-3} N/cm^2	
Shear strength = 138.28×10^{-3} N/cm^2	
Adhesive power	9.36×10^{-4} N/cm^2

2 Lime

Lime bought from the market was crushed, pulverized and sieved to obtain a powdered material of appropriate size distribution. The mechanical behavior and flow characteristics of the powdered lime are listed in Table 1.

3 Fluidization experiments

Fluidization experiments were performed with lime of +100 meshes and −120 meshes. Narrow particle size distribution was used because lime of that particle sizes is not liable to be carried away by the gas and the bed weight can thus be kept near its initial value.

Theoretically, the bed pressure drop increases with increase in gas flow rate until a critical fluidization velocity U_{mf} has been reached. When a material is fluidized, the bed pressure drop will keep constant, but the actual pressure drop of lime changes slightly with the gas flow. This is perhaps due to the occurrence of some channeling or slugging in the bed. Nevertheless, compared to Fisher-Tropsch catalyst, the deviation of lime is rather small. Taking 6 sets of data obtained with bed weight from 50 to 150g, the average deviation is only about 1%.

The pressure drop versus gas flow experiments show that, for this reactor, the appropriate bed weight is 50—150g and the gas velocity 0.15—0.4 m/s. Under these conditions, the gas rises up in the bed as bubbles which is typical for a bubbling-bed fluidization.

4 Attrition of lime particles in the reactor system

In a fluidized-bed reactor system, lime particle moves up and down in the reactor. When it collides with the reactor wall or with the cyclone of the reactor system, its particle size undergoes some changes caused by: (1) a grinding mechanism resulting in a decrease in particle diameter and emergence of some finest particles; (2) a disintegrating mechanism resulting in the emergence of several smaller particles with an over-all size distributions in conformity with certain math relations.

Both mechanisms have been found in industrial practices. The share of each is dependent upon reactor structure, operating conditions and particle behavior (Chen, 1995; NPRA, 1976; Wei, 1977; Feng, 1989; Vaux, 1985).

In this study, a lime of minus 160 meshes was used. The particle size distribution listed in Table 2 shows that most of the lime particles fall within the range of 65—98 μm. The mean particle diameter is 72.74 μm.

Table 2 Particle size distribution of lime

Size range, μm	0—65	65—76	76—90	90—98	98—100
Magnitude of diameter range Δd, μm	65	11	14	8	2
Mean diameter, d, μm	32.5	70.5	83	94	99
Weight% ΔR, %	7.77	23.64	50.73	16.81	1.05
Weight % per μm, $P=\Delta R/\Delta d$	0.12	2.15	3.62	2.10	0.53
Accumulated wt % retained on sieves	100	92.23	68.59	17.86	1.05

Fluidized-bed reactor system attrition tests were conducted at different bed weights and gas rates. During the experiments, lime collected in the 1st cyclone was recycled to the reactor, while that collected in the 2nd cyclone was discharged from the system. After 1 hour of fluidized attrition, the particle size distribution was measured again.

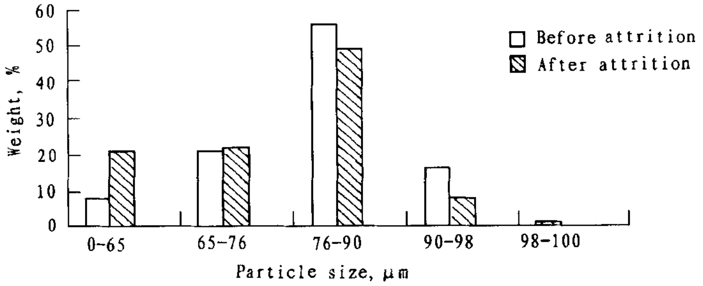

Fig.2 Particle size distribution before and after the attrition

Fig.2 shows that, as a result of attrition, the share of the 0—65 micrometer size fraction obviously increases, while those of all the bigger size fractions have different degrees of reductions. This shows that the attrition of bigger particles results in the emergence of a smaller particle and fines, instead of splitting into medium-sized ones. This is in conformity with the grinding mechanism mentioned earlier.

The rate of attrition can be calculated from the particle size distributions before and after the attrition. It is expressed as the decrease of particle diameter per unit time, i.e., μm/h. The rate of attrition increases with increase in gas velocity and decreases with increase in bed weight (Fig.3).

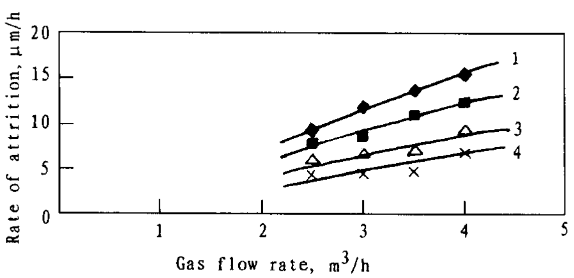

Fig.3 Rate of attrition versus bed weight and gas velocity
1. bed weight = 50; 2. bed weight = 80; 3. bed weight = 120; 4. bed weight = 150

Their relation can be expressed as follows:

$$B_r = KQ,$$

where B_r is the rate of attrition, μm/h; Q gas flow rate, m^3/h; K is a coefficient dependent on the bed weight, in $\mu m/m^3$; $K = 5.0 - 2.33 \times 10^{-2} W$; W is the bed weight, g.

The rate of attrition decreases with increase in time. This can be explained by the grinding mechanism which says that, as time goes by, the particle appears to be more and more spherical in shape resulting in less and less attrition.

With a bed weight of 80g, gas flow rate 3 m^3/h, the change of the rate of attrition with time is shown in Fig.4.

These relations are helpful to the development of a math model for the fluid-bed reactor system.

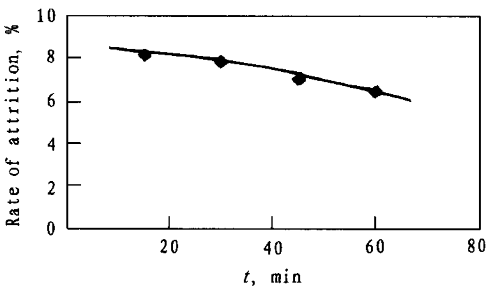

Fig.4 Rate of attrition versus time

5 Entrainment from the fluid-bed

Entrainment of lime particles from the fluid-bed is a rather complicated problem. Still there are not satisfactory theory and exact formula to describe this process. Even some basic view points are under dispute (Li, 1988; Kunii, 1990; 1991; Fournal, 1973). However, it is commonly agreed upon that the entrainment decreases with increase in the freeboard of the reactor and finally becomes stable. Zenz has put forward a TDH (theoretical disengaging height) idea which means the freeboard necessary to make the entrainment approaching a stable value.

A math equation has been derived from Zenz's experimental data. Substituting the max. gas velocity in this experiment (0.4 m/s) and the reactor diameter (0.06m) in the formula, the calculated TDH is about 0.4m. Since the gas outlet is 0.92m above the gas distributor, and the bed height is usually 0.3m, the actual freeboard is about 0.2m higher than the calculated TDH.

Terminating velocity of a particle is dependent on its particle size. For spherical particle, it can be calculated with a well-known formula. The terminating velocity of non-spherical particle (with the same equivalent diameter) is less than that of spherical ones. Using lime of particle size distribution as listed in Table 2, the lime collected in the 1st and 2nd cyclones, when the bed weight is 120g, gas flow 2.5—4.0 m^3/h and fluidization duration 10 min, had particle size distribution shown in Table 3.

Table 3 Particle size distribution of entrained lime (in wt %) vs gas flow rate

Gas flow rate, m^3/h	0—50μm	50—65μm	65—76μm	76—90μm	90—98μm	98—100μm
2.5	37.5	16.9	17.1	25.3	3.1	0.1
3.0	23.5	14.4	22.9	34.5	4.4	0.3
3.5	8.3	18.4	25.6	44.1	3.5	0.1
4.0	5.1	8.5	22.9	51.4	11.3	0.8

With these particle size distributions, the mean particle diameters have been calculated respectively and compared with the theoretical diameter of spherical ones under the same gas velocity. The results are shown in Table 4. It can be seen that the actual d_p and the theoretical ones have an average deviation of 5.2%, due to non-spherical characteristics of the lime particles.

Table 4 Particle size of entrained lime vs theoretically calculated diameters of spherical particles

Gas flow rate, m^3/h	Theoretical $d_{p'}$, μm	Actual d_p, μm	Deviation $K = d_p/d_{p'}$
2.5	41.17	42.10	1.022
3.0	49.41	50.52	1.022
3.5	57.64	63.10	1.095
4.0	65.84	70.24	1.067

6 Mass flow and particle size distribution models

A cold-modeling study of the circulating fluid-bed reactor system was conducted. The goal is to determine the steady-state mass flow and particle size distribution of the system and compare with values calculated by use of a math model developed from the previous individual studies.

6.1 The experiments

Most of the lime collected in the 1st cyclone was recycled to the reactor, a small part of it was discharged, lime collected in the 2nd cyclone was sent out of the system: some of the bed lime was continuously discharged from the reactor to keep the bed weight stable.

A steady state was reached after 120 min of operation.

6.2 Math model

A math model of mass flow rate and particle size distribution was developed, using data obtained in the attrition and entrainment experiments.

Given the feed rate, the bed discharge rate, the 1st cyclone collection efficiency and the recycle ratio of the 1st cyclone, a set of mass balance equations had been established, and the lime flow rates entering or leaving each equipment calculated. The recycle ratio is an arbitrary value. The smaller it is, the higher the load of the 1st cyclone will be. To avoid excessively heavy load, a suitable value, in consideration of the performance and efficiency of the cyclone being used, should be used.

Particle size distribution of the bed is an important factor for fluidized-bed reactors. It influences the flow behavior of the bed particles, the quality of fluidization, the bed weight and the entrainment loss. The lower limit of the particle size of the bed material depends on the cyclone performance, while the upper limit is dependent on the particle size distribution of the feed. Attrition behavior of the material also influences the bed particle sizes. A comparatively stable bed particle size distribution is crucial for keeping normal operation of the fluid-bed reactor.

Employing the previous findings of attrition and entrainment, a bed material particle size distribution model has been developed.

The general formula for the mass balance of each size range of particles is:

$$F_0P_0(R_i - nB_r)\Delta R_{i-n} - W(1 - \varepsilon)P_w(R_i - nB_r)\Delta R_{i-n} - F_3P_w(R_i - nB_r)\Delta R_{i-n} \\ - WP_w(R_i - nB_r)\Delta R_{i-n} + WP_w[R_i - (n - 1)B_r]\Delta R_{i-n+1} - WP_w[R_i - (n - 1)B_r]\Delta R_{i-n+1} \\ \times \{P_w[R_i - (n - 1)B_r][(R_i - nB_r)/(R_i - nB_r + B_r)]^3B_r\} = 0,$$

where F_0 is the feed rate in g/h; F_3 the discharge rate of the bed lime in g/h; W the bed weight in g; R the average diameter in μm; P the weight percent of a particular size range of particles whose average diameter is R; ΔR the weight percent per μm of that size range of particles; ε the recycle ratio of the 1st cyclone; B_r the rate of attrition in μm/h; suffix o denotes the feed; w denotes the bed; n denotes the numerical order of the mass balance equations. For that of the coarsest range of particles, $n = 0$; for that of the second coarsest range, $n = 1$, etc; i denotes the total number of size ranges.

Given F_0, F_3, W, ε and particle size distribution of the feed, the material balance of each size range of particles can be calculated successively until the particle size reaches 10 μm. Particles smaller than that can be considered not existing in the bed.

A computer program has been worked out to accelerate the calculation.

When F_0 is 60 g/h, W is 60g, w is 0.3 m/s, F_3 is 31.2 g/h, θ is 63.4%, ε is 83.2% and particle size distribution of the feed is as listed in Table 2, the calculated values and the experimental ones are shown in Table 5.

Table 5 Calculated flow rates and particle size distributions vs the actual values

	Calculated values	Actual values
Flow rate of lime entering 1st cyclone, g/h	78.7	NA
Flow rate of lime entering 2nd cyclone, g/h	14.2	13.2
Flow rate of lime discharged from the reactor, g/h	14.6	15.6
Mediated average particle diameter of the bed lime, μm	78.4	77.4

Comparison shows that the math model is basically in conformity with the experimental practice. It can therefore be used to predict what will happen if there is any change in the operating parameters of the system.

In this study, the retention time of lime in the reactor is about 1 hour, a value thousands longer than that in an entrained-bed reactor. The attrition rate is about 10 μm/h which will surely help remove the reaction product layer from the lime surface and promote the desulfurization reaction.

7 Proposed process flow scheme

In the cold-modeling experiments, lime has been found to have its own attrition characteristics. In an earlier experiment with sand, the rate of attrition increases with an increase in bed weight. Thus, at a given gas velocity, when the sand attrition is less than expected, the bed weight will gradually build up resulting in an increase in attrition rate, so the bed weight will finally reach a balanced state. But the case is entirely different for lime. So, when a circulating fluidized-bed reactor of lime is operated under a low gas velocity and low attrition rate, bed discharge is necessary to avoid excessive build-up of bed.

When the gas velocity is much higher than the terminating velocity of the coarsest particles in the feed, bed discharge is not necessary. But when water is sprayed into the bed to promote the reactivity of lime, agglomeration of bed lime might occur. So, even for this kind of circulating fluid-bed reactor, a bed discharge would be helpful to remove agglomerates from the bed and ensure smooth operation of the fluid-bed.

An elutriation system might be useful to recover fine particles and recycle them to the reactor.

8 Conclusions

Fluidization experiments showed that, for lime with particle size of 100 to 120 meshes, the appropriate conditions for fluidization are: gas velocity 0.25—0.4 m/s, bed weight 18—53 kg/m^2. Under these conditions, the bed pressure drop is 20—53 mm H_2O.

Attrition experiments showed that grinding is the major mechanism controlling the reduction of particle sizes during attrition. The rate of attrition increases with the rise in gas velocity, but decreases with the increase in bed weight. For the latter reason, the bed loses a self-balancing mechanism. To avoid excessive build-up of the bed, discharge of some of the bed lime seems to be necessary.

Entrainment experiments showed that the particle size distribution of the entrained lime is directly related to that of the feed. The mean diameter of the entrained particles is slightly bigger than the calculated diameter of spherical particle at the given gas velocity.

Continuous feeding, recycling, discharging of lime and steady-state bed weight and bed particle size distribution have been reached. The retention time of lime in the reactor was about 1 hour, and the rate of attrition was about 10 μm/h. These clearly shows the advantages of this kind of reactor.

A mathematical model of the reactor system has been developed which can be used to calculate the mass flow rates of lime particles and particle sizes distribution of the bed lime. Comparison showed that the calculated values were basically in conformity with the experimental ones.

As a result of this study, a flow scheme for desulfurization experiments has been proposed with a discharge system of bed lime when necessary.

Due to limitation of experimental conditions, the cold-modeling study was operated at a low gas velocity. It is proposed to employ a much higher gas velocity, say, 2—3 m/s, in the desulfurizaiton experiments to raise the productivity of the reactor and improve the process economics.

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