Advanced Powder Technology 25 (2014) 472–475

Contents lists available at SciVerse ScienceDirect

Advanced Powder Technology

journal homepage: www.elsevier.com/locate/apt

Rapid Communication

A study of the ash production behavior of spent limestone powders in CFBC

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ARTICLE INFO

Article history:
Received 7 February 2013
Received in revised form 19 April 2013
Accepted 23 April 2013
Available online 9 May 2013

Keywords:
Limestone
CFBC
Pore size distribution
Ash
Attrition

ABSTRACT

Spent limestone particles in a circulating fluidized bed combustor (CFBC) may show a bottom-ash-pro (BAP) behavior or a fly-ash-pro (FAP) behavior. In this study, two spent limestone particles showing the BAP behavior and two spent limestone particles showing the FAP behavior in a 125 MW CFBC are characterized based on their total pore volume and specific surface area before and after 800 °C calcination. Provided that the limestone primary fragmentation does not significantly affect the particle size distribution immediately after CFBC injection, the limestone particles having a relatively porous structure after 800 °C calcination show a BAP behavior in a CFBC.

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1. Introduction

A circulating fluidized bed combustor (CFBC) firing coals or petroleum cokes is commonly used as a steam generator or a power generator in industry. While firing coals or petroleum cokes in a CFBC, SO_2 emissions are usually *in situ* captured by calcium-based sorbents to avoid the air pollution. Limestone is one of the most commonly used calcium-based sorbents for the CFBC *in situ* desulphurization. The limestone particles are calcined, react with sulfur dioxide and oxygen to form calcium sulfate [1,2]:

$$CaCO_{3(s)} \rightarrow CaO_{(s)} + CO_{2(g)} \tag{1}$$

$$CaO_{(s)} + SO_{2(g)} + \frac{1}{2}O_{2(g)} \rightarrow CaSO_{4(s)} \tag{2}$$

The fully/partially sulfated powders are continuously removed from the top and the bottom of CFBC as the fly ashes and the bottom ashes, respectively. The size of the injected fresh limestone particles is usually much larger than the sizes of the fly and bottom ashes. For example, a 125 MW CFBC in Taiwan fires petroleum cokes and uses limestone particles with a mean size of 450 μm for *in situ* desulphurization. The mean particle sizes of the bottom ashes and of the fly ashes are 230 μm and 25 μm, respectively. The finer fly ashes are usually removed from the CFBC at the back-end with a bag-house and the coarser bottom ashes are usually removed from a CFBC by the screw transporter located at the bottom of the riser.

The limestone particle size distribution in a CFBC changes through particle attrition after the initial limestone particle primary fragmentation [3,4]. Previous studies showed that the limestone particle primary fragmentation occurred as a consequence of the thermal shock and the internal overpressure caused by carbon dioxide generation [3,5]. Particles over 200 μm can have residence times of the order of 10–20 h in an industrial CFBC. Nevertheless, the limestone particle primary fragmentation occurs within the first few minutes after the limestone particles injecting into the CFBC [1,4]. The limestone particle primary fragmentation determines the *initial* particle size distribution and particle attrition determines the *ultimate* particle size distribution in CFBC desulphurization. After the initial limestone particle primary fragmentation, the particle size distribution of the limestone particles in a CFBC is a function of the circulating time due to a much slower attrition size reduction mechanism.

Therefore, the ultimate particle size distribution of the spent sorbents in a CFBC is affected by the particle fragmentation and attrition mechanisms. These mechanisms influence the ratio of the fly ash production to the bottom ash production in a CFBC since the mean sizes of the fly ashes and bottom ashes are very different. The bottom ashes in a CFBC are removed by a screw transporter, the capacity of the screw transporter determines the maximum bottom ash removal rate. If the bottom ash production rate is much greater than the capacity of the screw transporter, the spent sorbents accumulate in a CFBC. The increasing of the solids holdup

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http://dx.doi.org/10.1016/j.apt.2013.04.014

in the system causes the increasing of the pressure drop across the bed. Defluidization may occur and the accumulation of the heat of combustion may lead to explosion [6].

It is therefore of practical importance to know whether the limestone powders favor the fly ash production or the bottom ash production in CFBC. Nevertheless, the limestone particles for CFBC desulphurization are usually characterized based on their reactivity in sulfation reactions [7], the fly-ash-pro (FAP) production behavior or the bottom-ash-pro (BAP) production behavior of the limestone particles is not considered. In this study, two limestone particles showing BAP behavior in a 125 MW CFBC (namely BAP limestones) and two limestone particles showing FAP behavior in a 125 MW CFBC (namely FAP limestones) are characterized based on their total pore volume and specific surface area before and after calcinations. The reasons for limestones showing the FAP production behavior or the BAP production behavior are also given.

2. Experimental methods

H, G, U and M limestones are used to study the ash production behavior in a 125 MW CFBC. They are from Guangdong, Guangxi, Liaoning and Liaoning in China, respectively. The true particle densities and the mass fractions of the major components of four limestones are shown in Table 1. These limestones have been used for *in situ* desulphurization in a 125 MW full-scale CFBC in Taiwan. The operating conditions are as follows. The limestones were pulverized and the powders of the size less than 710 μm were injected into the CFBC at the rate of 26 ton h^{-1}. The petroleum cokes were fired at the rate of 41 ton h^{-1} in the CFBC and the steam is generated at the rate of 450 ton h^{-1}. The temperature of the CFBC was 900 °C and the air superficial velocity was 5.18 m s^{-1}. When injecting U and M limestones for desulphurization, the ratio of the fly ash production rate (on the mass bases) to the bottom ash production rate was 7:3. When injecting H and G limestones for desulphurization, the ratio of the fly ash production rate to the bottom ash production rate was 2:3. Therefore, U and M limestones show FAP behavior and they are FAP limestones. H and G limestones show BAP behavior and they are BAP limestones.

In the lab tests, the limestones were pulverized and the powders with the sieving size of 105–149 μm were used in the calcination experiments. 10.0 g pulverized limestone powders were calcined in a tubular oven. The oven was heated from room temperature to 800 °C at the heating rate of 10 °C min^{-1} and maintained at 800 °C for 5 h. The powders before and after calcination were characterized using X-ray diffraction (XRD, Bruker-AXS D5005D, Siemens). The total pore volume and the specific surface area of the powders before and after calcination were determined by the BET method using nitrogen as the adsorption gas (ASAP 2020, Micromeritics).

3. Results and discussion

Fig. 1a and b show the XRD spectra of the powders before and after calcination, respectively. While all limestones show calcium carbonate calcite peaks in Fig. 1a, H powder shows extra dolomite ($CaMg(CO_3)_2$) peaks. Dolomite accompanied calcium carbonate is frequently found in CFBC desulphurization application. After calcination, calcium carbonate produces calcium oxide and dolomite produces calcium oxide and magnesium oxide. The corresponding peaks of calcium oxide and magnesium oxide are observed in Fig. 1b. Except for dolomite found in limestone H, the FAP limestones and the BAP limestones do not show apparent differences in XRD spectrums.

The comparisons of the total pore volume and the specific surface area of four limestones before and after calcination are shown in Table 2 and Fig. 2a, and Table 2 and Fig. 2b, respectively. Carbon dioxide was produced and hence the powder total pore volume and the specific surface area were significantly increased after calcination in all cases. However, it is interesting to note that the total pore volume and the specific surface area of four powders before and after calcination can be classified into two groups. The group classification is corresponding to the BAP/FAP classification. The FAP limestones, U and M, show greater specific surface areas before calcination and less carbon dioxide generation per unit mass of

Table 1
The true density and the mass fraction of the major components of each limestone.

Limestone	$CaCO_3$ (wt%)	SiO_2 (wt%)	Moisture (wt%)	Balance (wt%)	True density (g cm^{-3})
H	93.28	2.55	0.688	3.482	2.7774
G	95.24	3.86	0.607	0.293	2.7399
U	92.90	1.64	0.065	5.395	2.7569
M	96.31	1.21	0.046	2.439	2.7501

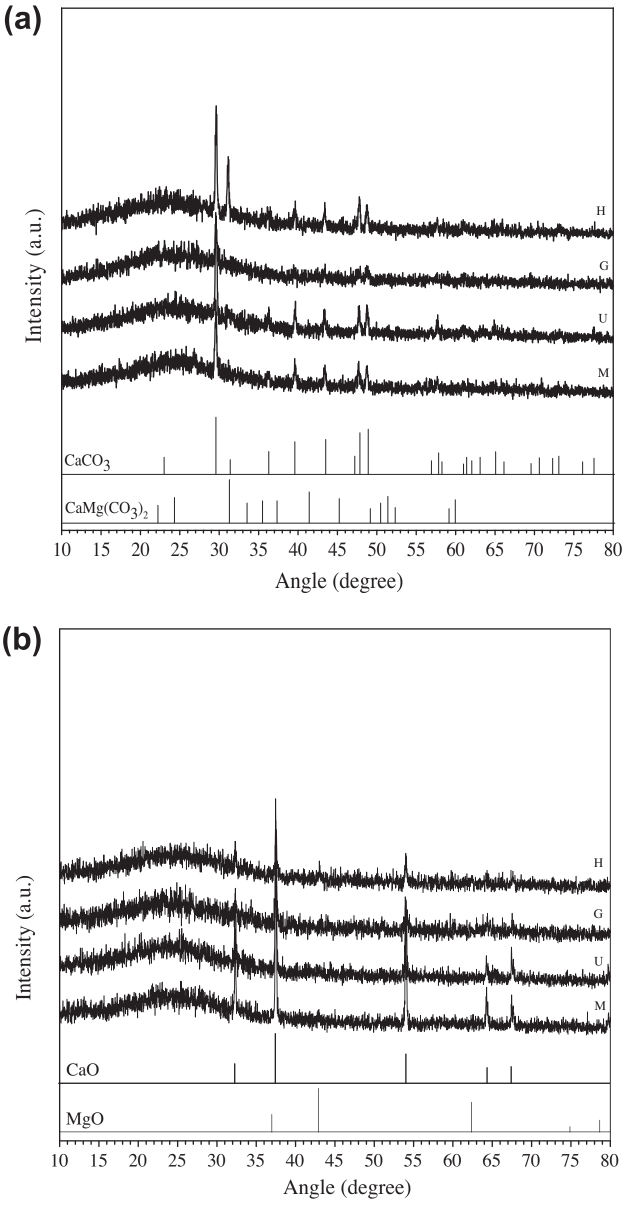

Fig. 1. XRD spectra of the powders: (a) before calcination and (b) after calcination.

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