C-4.2 Control Techniques of SOx and NOx in East Asia and Their Evaluation

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Abstract This report describes the control methods of SO_2 and NOx from coal combustion, which cause acid rain in the east asia, for developing countries. Desulfurization characteristics of some kinds of Chinese limestones were examined. Desulfurization capability of Chinese limestone was as well as that of Japanese one.

Direct injection of fine absorbent such as limestone, dolomite and limehydrate to the pulverized coal combustor was tested. Up to 80% of sulfur removal was achieved when limestone was injected at Ca/S = 4. Limehydrate showed the highest desulfurization capability among the tested absorbents. Sulfur removal was not extreamly avoided when the two staged combustion technique was adopted to reduce NO emission.

Circulating fluidized bed combustion (CFBC) was tested. NO emission from CFBC was about 100 ppm for typical Chinese coal.

Key Words Acid rain, Coal combustion, Desulfurization

1. Introduction

Large amount of SOx and NOx emission in east Asia region causes acid rain. The main sources of above SOx and NOx are stationary coal combustion facilities. In particular, China depends on coal as main energy sources. In 1988, 76% of China's primary energy demand was covered by coal combustion. As some Chines coals produced in the south western part have very high sulfur contents, acid rain becomes very serious environmental problem in the south western part of China in recent years.

One of the most effective methods to prevent acid rain is the addition of flue gas desulfurization facility to conventional coal combustors. It is, however, very difficult to adopt a desulfurization facility to each coal combustor, because construction and running cost are high and the maintenance is difficult for developing countries. These countries require the technology that is easy to use and low cost. In recent years, cheaper and easier desulfurization technologies for the developing countries are newly developing. The direct injection system of absorbents and the circulating fluidized bed combustion are these technologies. In this research program, the direct injection of absorbents to the pulverized coal combustor and CFBC technology were studied.

2. Research Objective

The concept of the direct injection desulfurization is that the fine powder of absorbents such as limestone, limehydrate and dolomite is directly injected into the boiler furnace and remove SO_2 by the fixation reaction between absorbent and SO_2 in flue gas, which produces calcium sulfate and/or magnesium sulfate. At first, the desulfurization characteristics of Chinese limestones were examined in comparison with Japanese one. In the research concerning with the direct injection of absorbents, the research objective is to clear up the optimal operating conditions. Effects of quantity, size distribution, chemical composition and injection points on SO_2 removal were examined. Furthermore, effect of two staged combustion on direct desulfurization was tested.

In CFBC research work, NO emission level was mainly examined.

3. Experiment, results and discussion

3.1 Desulfurization characteristics of Chinese limestones

Desulfurization characteristics of three kinds of Chinese limestone (Dou Fong Dong, Nang Dian and Da Ming San) and one Japanese limestone (Kansuiseki) were tested using a batch reactor. Small amounts of limestone particles were mixed with inert quartz sand and formed fixed bed, which was heated up to 1100 K and $SO_2/O_2/Ar$ gas mixture was passed through the fixed bed. Outlet SO_2 concentration was monitored continuously. Typical outlet SO_2 concentration changes with time are shown in Fig.1. Sulfation reaction was saturated within 30 min. Captured SO_2 was calculated by integration of these SO_2 vs. time curves. Figure 2 shows the SO_2 captured for each limestone. Dou Fong Dong limestone shows the largest absorbing capacity and Nang Dian limestone shows the smallest among the tested limestones. Reaction capacity of Dou Fong Dong limestone is as well as that of Kansuiseki limestone.

Figure 3 shows the pore structure of Dou Fong Dong and Nang Dian limestone. Obviously, the

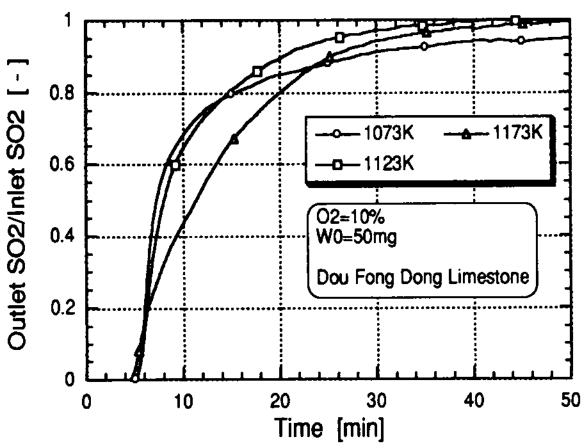


Fig. 1 Typical outlet SO_2 concentration change with time.

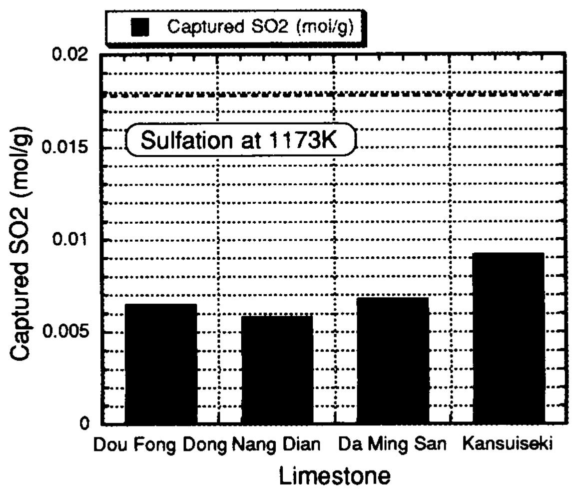


Fig. 2 SO_2 capture at 1173K.

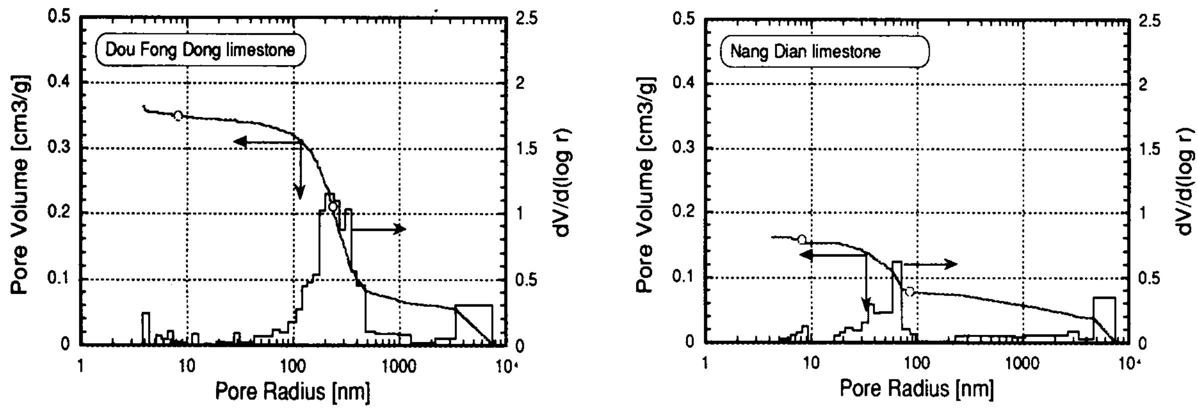


Fig. 3 Pore structure of Dou Fong Dong and Nang Dian limestone.

pore structure is well developed for Dou Fong Dong limestone in comparison with that of Nang Dian limestone. Total pore volume and surface area of Dou Fong Dong is larger than those of Nang Dian limestone. This results shows that the reactivity in the desulfurization reaction is closely connected with the pore structure after calcination.

3.2 Direct injection of absorbents into the pulverized coal combustor

The direct injection desulfurization system was carried out using a pulverized coal combustion system. Figure 4 shows the schematic diagram of an experimental system. The combustion chamber is 0.4m in inside diameter and 4m in height, and its capacity is 10kg-coal/hr. There are eight ports for observation and injection of staging air and/or absorbents. The concentration of SO_2, NO, CO_2, CO and SO_2 in the flue gas was continuously monitored.

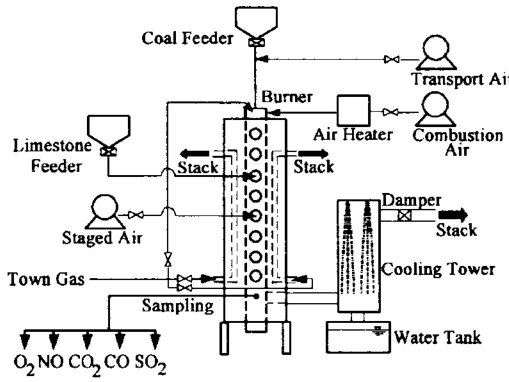


Fig. 4 Schematic diagram of an experimental pulverized coal combustor.

Miike coal, Japanese domestic coal with high sulfur content of 1.7%, was fired to simulate high sulfur

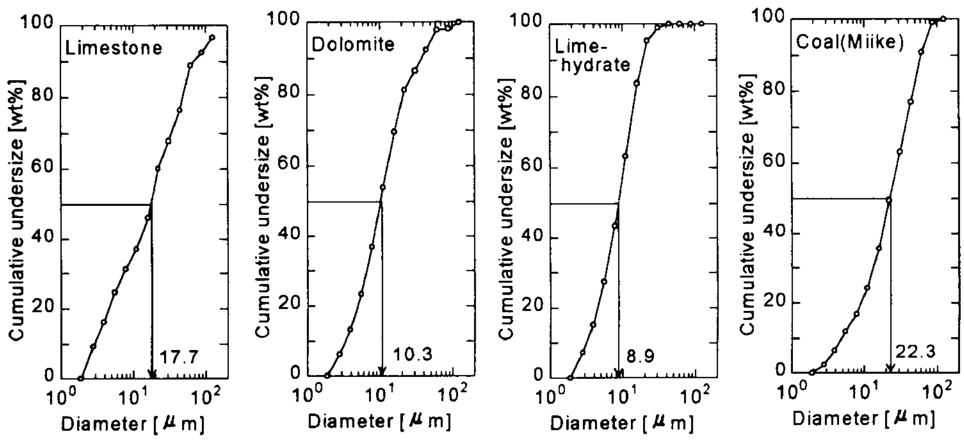


Fig. 5 Size distribution of coal and absorbents.

coal used in China. Limestone was used as the main absorbent, because it is easily obtained. To compare the desulfurization efficiencies, limehydrate and dolomite are also used. Figure 5 shows the size distribution of coal and absorbents.

Figure 6 shows the relation between Ca/S molar ratio and the SO2 concentration in flue gas. SO_2 concentration decreases with increasing Ca/S. Up to 80 % of the desulfurization efficiency is achieved at 4 of Ca/S.

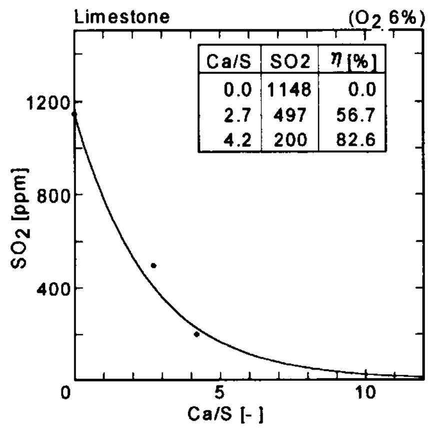


Fig. 6 Effect of limestone injection.

Limehydrate shows the highest desulfurization efficiency. Figure 7 shows the comparison among different types of absorbents.

The temperature of injection point is regarded as the most significant factor to desulfurization. Figure 8 shows the temperature of injection points and the SO_2 concentration in flue gas. On this system, the most effective temperature was about 1400 K.

As the desulfurization reaction requires O_2, SO_2 removal is usually reduced when the two staged combustion is adopted. Figure 9 shows the SO_2 and NOx concentration as a function of staging. NOx is remarkably reduced by the two staged combustion and the desulfurization efficiency is kept as almost constant. This means that the two staged combustion has small effect on desulfurization by the direct injection.

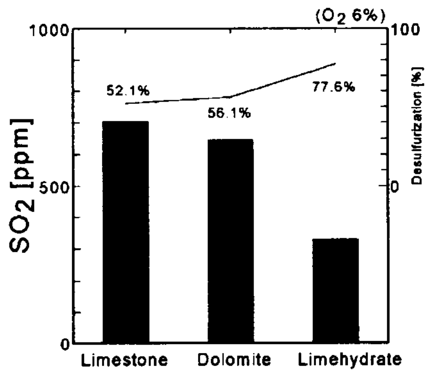


Fig. 7 Comparison among absorbents.

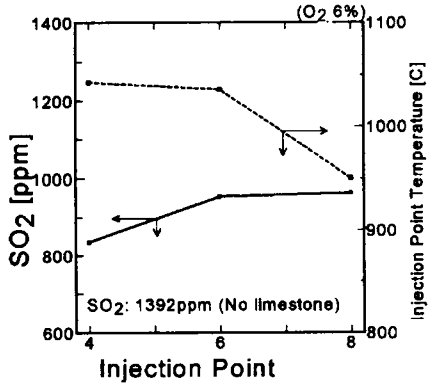


Fig. 8 Effect of injection points on SO_2 removal.

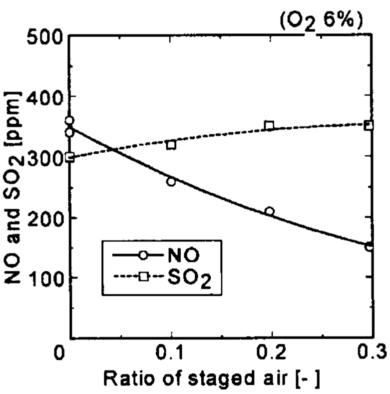


Fig. 9 NO and SO_2 emissions during

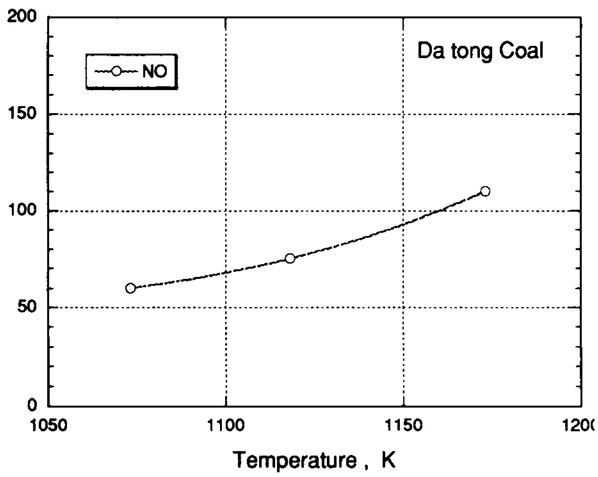
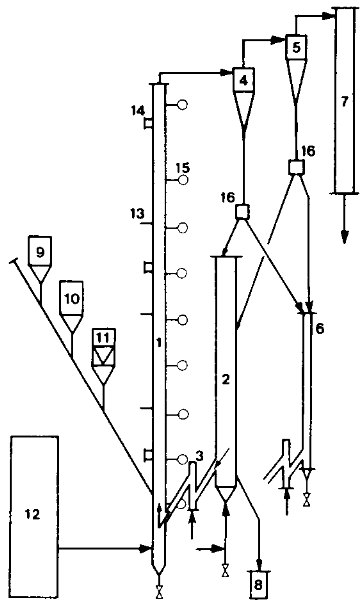


Fig. 11 NO emission from bench scale CFBC for Datong coal.

3.3 Circulating fluidized bed combustion

Circulating fluidized bed combustion (CFBC) is one of the most advanced coal combustion system with a number of good points such as high flexibility to coal types, high combustion efficiency, high limestone utilization and low NOx emission. Typical Chinese coal (Datong coal) was fired by a bench scale CFBC, 100 mm inside diameter and 5000 mm height, illustrated in Fig. 10. Figure 11 shows the NO emission level for Datong coal. NO emission level was below 110 ppm in whole temperature range.



1 riser
2 downcomer
3 N-shaped pneumatic valve
4 primary cyclone
5 secondary cyclone
6 secondary downcomer
7 gas cooler
8 ash pod
9 limestone feeder
10 sand hopper
11 coal feeder
12 air preheater
13 air injection port
14 view port
15 thermocouples & pressure transducer
16 switching valve

Fig. 10 Bemch scale CFBC.

4. Conclusion

Through this study, we concluded as follows:

1) Desulfurization capability of Chinese limestone was as well as that of Japanese one.

2) The desulfurization efficiency could be achieved about 80% at 4 of Ca/S molar ratio. Limhydrate showed the highest desulfurization efficiency among the absorbents used in these experiments. On this system, the most high efficiency injection point temperature was about 1400 K. The NOx emission is remarkably reduced by the two staged combustion and the desulfurization efficiency by direct injection is not so influenced by staging.

3) NO emission from CFBC was about 100 ppm for typical Chinese coal.