Mechanism of flue gas simultaneous desulfurization and denitrification using the highly reactive absorbent

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Abstract Fly ash, industry-grade lime and a few oxidizing manganese compound additive were used to prepare the “Oxygen-riched” highly reactive absorbent for simultaneous desulfurization and denitrification. Experiments of simultaneous desulfurization and denitrification were carried out using the highly reactive absorbent in the flue gas circulating fluidized bed (CFB) system. Removal efficiencies of 94.5% for SO2 and 64.2% for NO were obtained respectively. The scanning electron microscope (SEM) and accessory X-ray energy spectrometer were used to observe micro-properties of the samples, including fly ash, common highly reactive absorbent, “Oxygen-riched” highly reactive absorbent and spent absorbent. The white flake layers were observed in the SEM images about surfaces of the common highly reactive absorbent and “Oxygen-riched” one, and the particle surfaces of the spent absorbent were porous. The content of calcium on surface was higher than that of the average in the highly reactive absorbent. The manganese compound additive dispersed uniformly on the surfaces of the “Oxygen-riched” highly reactive absorbent. There was a sulfur peak in the energy spectra pictures of the spent absorbent. The component of the spent absorbent was analyzed with chemical analysis methods, and the results indicated that more nitrogen species appeared in the absorbent except sulfur species, and SO2 and NO were removed by chemical absorption according to the experimental results of X-ray energy spectrometer and the chemical analysis. Sulfate being the main desulfurization products, nitrite was the main denitrification ones during the process, in which NO was oxidized rapidly to NO2 and absorbed by the chemical reaction.

Keywords: highly reactive absorbent, fly ash, simultaneous desulfurization and denitrification, micro-property, scanning electron microscope, X-ray energy spectra.

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Using slurry mixed by fly ash and alkaline material, removal of SO2 in industrial waste gas had been reported in the 1970s, which was named the wet scrubbing[1]. Since
then, there have been many reports about flue gas desulfurization using the reactive absorbent prepared with flying ash. There are four kinds as follows: (1) Using slurry of fly ash as absorbent\[^{2-4}\], which mainly utilize the property of fly ash for desulfurization, but the removal efficiency is not high. (2) Using fly ash and lime/slaked lime\[^{5-17}\] as absorbent. There have been a lot of reports about the preparation conditions of this kind of absorbent, such as digesting temperature, time, pressure ratio by weight of fly ash and calcium hydroxide, and ratio of water and solid, etc.\[^{6-9,11,15}\]. (3) Using highly reactive absorbent. The additives, such as CaSO\(_4\)\[^{18-24}\], CaCl\(_2\) and other compounds\[^{19}\], are added to the fly ash/lime to improve the desulfurization activity. Unfortunately, the mentioned absorbents are only used for desulfurization, while simultaneous denitrification cannot be realized. The “Oxygen-riched” absorbent for simultaneous desulfurization and denitrification was prepared by adding oxidative reagent to fly ash and lime. With the duct injection system and CFB as the working platform, the experiments about simultaneous removal of SO\(_2\) and NO in flue gas have been done by using the proposed absorbent, which are reported in our papers\[^{25-27}\]. The “Oxygen-riched” absorbent is named the fourth kind.

The present investigations of the former three kinds of absorbents showed that the surface characteristic of the absorbents has a great effect on desulfurization. However, the surface characteristics of the fourth kind of absorbent and its reactions with SO\(_2\) and NO in the CFB system have not been reported. SEM and the accessory X-ray energy spectrometer were used to analyze micro-properties and contents of significant elements on the surface of the fly ash, the fly ash/lime absorbent, “Oxygen-riched” absorbent and reaction products in this paper. The chemical analysis methods were adopted to determine the compositions of the products of desulfurization and denitrification. Thus the detailed mechanism of desulfurization and denitrification in the CFB system was investigated.

1 Preparation of the “Oxygen-riched” highly reactive absorbent

According to our previous experimental results\[^{25,26}\] and other research conclusions\[^{13,17,28}\], the “Oxygen-riched” highly reactive absorbent was prepared from fly ash, industrial lime and oxidizing manganese compound additive. The preparation process was as follows. The mixture including fly ash and industrial lime with the ratio of 3:1 in weight, manganese compound and water, was stirred and digested in 363 K and dried after six hours. Thus, the “Oxygen-riched” highly reactive absorbent was achieved.

The fly ash used in the experiments came from Baoding Thermoelectricity Plant, whose compositions are shown in Table 1. The content of the effective calcia in the industrial lime was 90.77%, measured by the method of cane sugar.

<table>
<thead>
<tr>
<th>Components</th>
<th>SiO(_2)</th>
<th>Al(_2)O(_3)</th>
<th>Fe(_2)O(_3)</th>
<th>CaO</th>
<th>MgO</th>
<th>Combustion loss</th>
<th>Others</th>
</tr>
</thead>
<tbody>
<tr>
<td>Content (%)</td>
<td>46.6</td>
<td>25.2</td>
<td>11.1 3.3</td>
<td>2.4</td>
<td>10.0</td>
<td>1.4</td>
<td></td>
</tr>
</tbody>
</table>

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