CATALYTIC REDUCTION OF SO$_2$ UNDER REGENERATOR OFF-GAS CONTAINING OXYGEN OVER Cu-Sn-Zr BASED OXIDES FOR HOT COAL GAS DESULFURIZATION PROCESS

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Abstract:
In coal gasification process, sulfur compounds such as H$_2$S and COS can be produced, due to the sulfur component in coal, and the hydrogen sulfide contained in coal derived synthetic gas can be selectively treated using the hot gas desulfurization process with a desulfurization sorbents. An SO$_2$ is produced in the regeneration process of the sulfide sorbent. SO$_2$ can be removed by catalytic reduction process using a reducing agent. However, the regeneration gas exhausted in the regenerator of hot gas desulfurization process contained the low content of unreacted O$_2$, which is the remained oxygen in the re-oxidation of metal sulfide used as the desulfurization sorbent. Since, this oxygen can leads an oxidation of the elemental sulfur, the conversion of SO$_2$ decreased due to the production of SO$_2$.

In order to find a suitable catalyst for DS RP (direct sulfur recovery), Cu-Sn-Zr based catalysts were prepared and their catalytic activity was examined in this study. In this study, the catalytic activity of Cu-Sn-Zr based catalysts was tested for SO$_2$ reduction under oxygen presence condition.

Copper based catalyst has the high activity for CO oxidation because copper well chemisorbed oxygen. Therefore, it could be expected that the unreacted oxygen exhausted from the regenerator is removed by CO oxidation over Cu-sites of Sn-Zr based catalyst promoted with copper. Meanwhile, Cu-sites in Sn-Zr based catalyst are well sulfidated to CuS and COS can be produced the gas-solid reaction between the produced CuS and CO. COS and SO$_2$ adsorbed on Lewis and Brønsted acid sites in ZrO$_2$, respectively, can be converted to element sulfur by Langmuir- Hinshelwood mechanism. This reaction path way is called to COS intermediate mechanism and the oxygen effect on the catalytic reduction of SO$_2$ be prevented. The catalytic activity tests for SO$_2$ catalytic reduction were carried out over Cu-Sn-Zr based catalysts prepared with changing content of Cu. The conversion of SO$_2$ and the yield of elemental sulfur increased with increasing Cu content in Cu-Sn-Zr based catalysts. It was concluded that the decreasing SO$_2$ conversion effected with the unreacted oxygen contained in regenerator off-gases was reduced by CO oxidation and COS intermediate mechanism with Cu sites over Sn-Zr based catalyst.

Keywords: coal gas cleaning, catalytic reduction, sulfur dioxide, COS intermediate

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