Integrated Removal of NOx,, with Carbon Monoxide as Reductant, and Capture of Mercury in a Low Temperature Selective Catalytic and Adsorptive Reactor

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A multi-year investigation is in progress to develop a novel, advanced Low Temperature Selective Catalytic and Adsorptive Reactor (LTSCAR) for the simultaneous removal of NO_x and mercury (elemental and oxidized) from coal-fired flue gases in a single unit. It is anticipated that the proposed system will lower removal costs for the targeted pollutants by an order of magnitude relative to current technologies.

This effort has resulted in the development of a novel low temperature SCR catalyst, $MnO₂$ supported on titania; this catalyst has been proven to be effective for elemental mercury capture at elevated temperatures (175-200 $^{\circ}$ C). Low temperature NO_x reduction with CO showed very high NO_x conversion; nearly 100% conversion was observed over 10 wt% $MnO₂/TiO₂$ at 200°C and industrially relevant conditions (50,000 $hr⁻¹$ space velocity, 2 vol % oxygen).

To investigate potential synergies between SCR and mercury adsorption, a sample of fresh catalyst was tested for use as a mercury adsorbent. Elemental mercury uptake as high as 17.4 mg/g was observed at 200 °C for mercury capture from a nitrogen carrier. The catalyst retained activity for mercury capture after use in NO_x reduction. The manganese loading and bed temperature, which influence surface oxide composition, were found to be important factors for mercury capture. X-ray photoelectron spectroscopy (XPS) results indicate that the $MnO₂$ in the catalyst acts as a reactant in the mercury removal process.

A parallel project task focused on elemental mercury removal by adsorption on a nanostructured chelating adsorbent at elevated temperatures. The adsorbent consists of a mesoporous silica substrate functionalized with groups that can capture mercury by chelation; the functionalized silica is coated with an ionic liquid to allow oxidized mercury to exist in an ionized form to facilitate chelation. The current effort focuses on identifying ionic liquids that can oxidize elemental mercury for subsequent immobilization by chelation. A range of oxidizing ionic liquids have been synthesized and screened for mercury capture characteristics and thermal stability. The ionic liquids screened to date – 1-butyl-1-methyl pyrrolidinium bis(trifluoromethane sulfonyl)imide (P_{14}) , 1-butyl-pyridinium bis(trifluoromethane sulfonyl) imide, and 1-butyl-3-methyl imidazolium bis(trifluoromethane sulfonyl) imide – all have the required thermal stability. P_{14} has the highest capacity for elemental mercury capture.

The results of this project to date support the feasibility of the core process concept that reduced SCR temperatures coupled with improved mercury capture performance at SCR temperatures can significantly improve the operation and cost effectiveness of environmental controls for coal combustion.