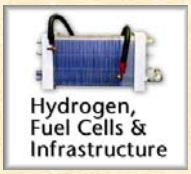
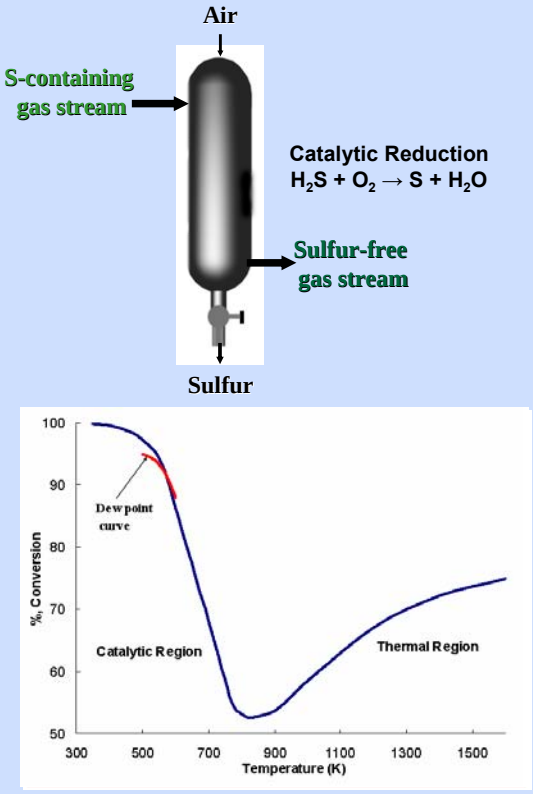


Carbon Catalysts Remove Sulfur to Clean Fuel Cell Feed Gases

Novel Activated Carbon Reduces Sulfur in Hydrogen Fuels to < 100 ppb

Background

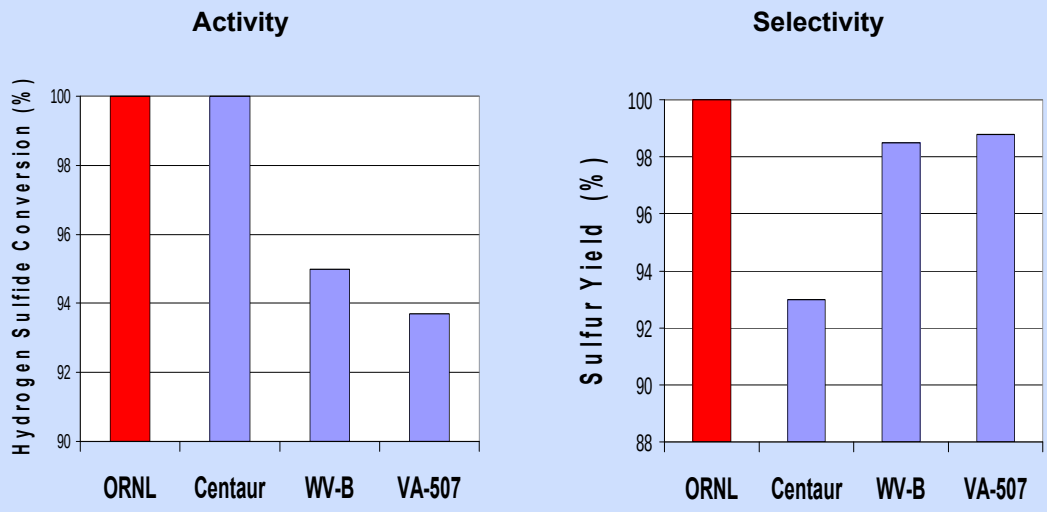
Proton exchange membrane (PEM) fuel cells can provide high efficiency power generation with near-zero emissions, but sensitivity of the fuel cell catalysts to impurities such as sulfur may require the use of high-purity hydrogen feedstocks. Sulfur in H_2 feed supplies poisons fuel cell catalysts, and even concentrations as low as a few ppm of hydrogen sulfide can dramatically shorten catalyst life. A process based on the selective catalytic oxidation of H_2S to elemental sulfur is a very attractive technology for eliminating sulfur poisoning. The reaction has the thermodynamic potential to remove H_2S to the parts-per-billion (ppb) level at temperatures below 200°C. Gaseous fuel is cleaned and elemental sulfur is recovered in a single-step process. The process works without consuming H_2 or other useful fuel components. The use of activated carbon as a catalyst for selective partial oxidation of hydrogen sulfide (H_2S) could be a preferred approach for the removal of H_2S from gas streams. Preventing the formation of unwanted gaseous sulfur by-products, such as sulfur dioxide (SO_2) and carbonyl sulfide (COS), is one of the main challenges.



The effective temperature range of reduction of H_2S to the ppb level extends to 500°K (~200°C)

Accomplishments

The use of carbon-based materials as catalysts for selective oxidation of H_2S in hydrogen-rich gas streams has been demonstrated. Several commercial and lab-made catalysts were evaluated catalytically in a fixed-bed reactor system with the concentration of the exhausted gases being monitored by a gas chromatograph (GC) and a Fourier transform infrared (FTIR) spectrometer. The inlet gas mixture was varied to include other common components found in a reformate stream, such as CO, CO_2 and H_2O. ORNL lab-made catalysts have demonstrated the capability of continuous removal of sulfur to levels below 100 ppb. The ORNL catalyst consistently reduced H_2S without formation of unwanted byproducts such as SO_2 and COS for more than 15 hours.



Selective catalytic oxidation of 1,000 ppm H_2S in H_2 in ten hours of stream at 150°C comparing three commercial catalysts (Centaur, WV-B, and VA-507) to the ORNL catalyst. Commercial catalysts either show a strong activity with low selectivity, or average activity with good selectivity. The ORNL catalyst exhibits high conversion (activity) with no by-products (selectivity).

Several properties such as precursor nature, impurity content and type, pore structure, and surface functionality have been found to contribute to the catalytic properties of the carbon materials. The catalytic activity of the carbons are correlated to their microstructure, the amount of oxygen-containing groups on the surface, and nitrogen content. Optimization of the activity and selectivity is being carried out by introduction of catalytically active nitrogen into the catalyst structure to further improve the adsorption capacity of this carbon for H_2S.

Benefits

- Low-cost catalyst
- Process operates at low temperatures
- No gaseous emissions - only solid sulfur produced
- Insensitive to steam in feed gas
- Eliminates both H_2S and COS

Future Work

One future goal is to extend the initial period in which H_2S is completely and exclusively converted to S, so that a complete elimination of sulfur compounds is achieved. This step will utilize optimization of synthesis process based on fundamental studies of the reaction mechanism and catalytic properties.

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