

Development of Sulfur Tolerant Catalyst for Jet Fuel Steam Reforming

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Introduction

Aircraft manufacturers are being required to reduce the emissions and lower the overall fuel consumption of their systems. Employing a fuel cell to power the electric needs on the aircraft is one way forward. To that end, NASA envisions employing fuel cells running on jet fuel reformates for its uninhabited aerial vehicles (UAV) and low emission alternative power (LEAP) missions, as well as for transatlantic and intercontinental flights. Additionally, the military foresees utilizing jet fuel, specifically JP-8, as a platform fuel in their fuel cell applications. The onboard jet fuel can have sulfur content as high as 3000 ppm, thus, a sulfur-tolerant reforming catalysts is required in order to use jet fuel as a hydrogen source for the fuel cell. The current presentation describes the development of a novel sulfur-tolerant reforming catalyst that is active and stable for conversion of jet fuel to hydrogen through steam reforming.

Materials and Methods

Catalysts were prepared from commercially available alumina supports on which active metal and stabilizers were supported via wet impregnation. Experiments were completed using a simulated jet fuel comprised of hexadecane (70%), toluene (15%), tetralin (10%) and methyl-naphthalene (5%). Several different organic species were evaluated as a source of sulfur to study the effect of sulfur poisoning. Steam reforming of this simulated mixture was studied at 800°C, a steam to carbon ratio of 3, and a GHSV of 80,000 hr⁻¹ at reaction condition. The product gases were analyzed using a Shimadzu GC-2010. Catalysts were characterized using X-ray diffraction (XRD), BET surface area and dispersion by pulsed chemisorption. Inductively coupled plasma (ICP) is used to identify sub ppm levels of sulfur.

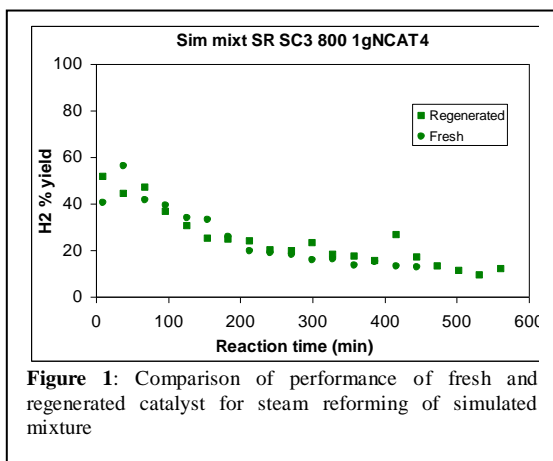


Figure 1: Comparison of performance of fresh and regenerated catalyst for steam reforming of simulated mixture

Results and Discussion

In our previous work, we demonstrated that catalyst NCAT4 gave good activity and stability with hexadecane steam reforming with up to 250 ppm of sulfur in the feed. Here, we demonstrate that during steam reforming of sulfur free simulated mixture, 60% hydrogen yield is initially obtained, although the activity drops steadily with time. Regeneration in air allows full recovery of the initial catalyst activity, as evident by similar performance of catalyst after regeneration, and shown in Figure 1. This result is

consistent with other analyses that suggest the deactivation is primarily from formation of coke from decomposition of methyl naphthalene.

Reaction of feed mixtures containing 100 ppm sulfur reveal a more dramatic decrease in catalyst activity than was observed with steam reforming of sulfur free simulated mixture. Figure 2, which describes the rate of hydrogen sulfide formation from experiments using different sulfur-containing species, indicates that all sulfur species produce H₂S in similar quantities, and that no sulfur-containing product is produced during the initial time-on-stream. This may suggest that the organosulfur compound is first adsorbed as a similar surface component independent of initial structure. Surface analysis of the catalyst following reaction shows the presence of adsorbed sulfur and increased levels of coke formation at higher sulfur loadings.

XRD studies were done with NCAT4 after steam reforming of simulated mixture doped to 100 ppm sulfur using thiophene and benzothiophene. Formation of graphitic coke was evident from its characteristic peak. We could not detect the presence of sulfur on the surface, likely because the amount of sulfur formed on the catalyst is below the detection limit of the XRD. ICP analysis is assumed to provide information about the amount of sulfur deposited on the catalyst and further analysis of the XRD data is underway to more fully understand the role of fuel sulfur on the catalyst surface.

Significance

Fuel cell auxiliary power units (APUs) using on-board hydrogen produced from liquid fuels are an attractive alternative as a source of electric power onboard aircraft. It is advantageous to produce hydrogen from on-board fuels as it avoids the storage issues associated with using pure hydrogen. The major challenge in jet fuel reforming arises because it contains a high percentage of sulfur, and the catalysts used for reforming are severely poisoned by the sulfur present. Several catalysts have been identified that show promise for their stability during jet fuel reforming, and analysis of the mechanisms of deactivation has been identified.

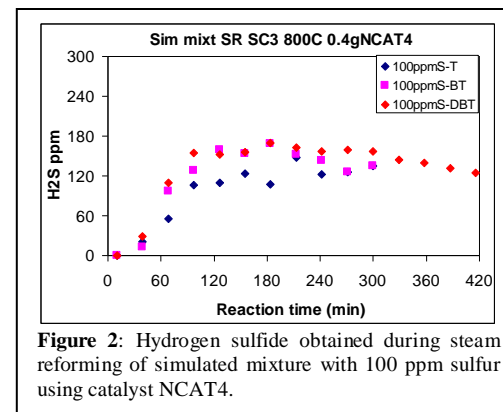


Figure 2: Hydrogen sulfide obtained during steam reforming of simulated mixture with 100 ppm sulfur using catalyst NCAT4.

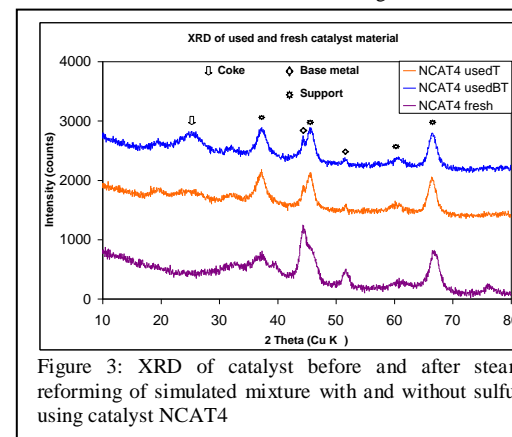


Figure 3: XRD of catalyst before and after steam reforming of simulated mixture with and without sulfur using catalyst NCAT4