

Bimetallic Catalyst Development for Dry Reforming of Methane and Hydrogenation of Succinic Acid

**Dissertation Defense by
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Abstract

Advances in natural gas recovery have greatly impacted the global economy; the abundant supply of gas has revealed new opportunities and challenges in a wide range of markets within the fuels and chemicals industries. One such opportunity exists with the overabundance of gas from remote wells that is flared or emitted because of the high costs associated with transportation of a chemical with such a high molar volume. A possible solution is an on-site process to convert gas to higher value chemicals that are liquid at standard conditions, and can more easily be transported. As a first step, CO₂ from the process may be utilized to convert CH₄, the main component of natural gas, to synthesis gas through dry reforming of methane (DRM). Synthesis gas, a mixture of H₂ and CO, may then be converted to liquid chemicals through a number of different reactions. In this study, we prepared a series of Pt-promoted Ni bimetallic catalysts by electroless deposition (ED) and evaluated them for DRM. An enhancement in activity was observed with the addition of an intermediate loading of Pt, which is associated with the formation of a NiPt alloy phase. However, the overall stability decreases with increasing Pt loading because of the high rate of carbon formation. Post-reaction characterization reveals phase separation of the alloy at high temperatures.

Another opportunity arises out of cheap natural gas as petrochemical producers shift from oil-based naphtha cracking to natural gas cracking for ethylene production. Since gas cracking produces none of the heavier hydrocarbon co-products that naphtha cracking does, the result is a growing gap in the supply of C₄ building block chemicals. One possible alternative is to utilize biomass-based compounds, such as succinic acid (SAC), which is a C₄-diacid. To reach many of the end markets that succinic acid is used as feed, it is necessary to hydrogenate it to downstream high-performance chemicals such as γ -butyrolactone (GBL), 1,4-butanediol (BDO), and tetrahydrofuran (THF). In this work, we evaluated a series of Ir-Re/C bimetallic catalysts prepared by strong electrostatic adsorption (SEA) for aqueous phase hydrogenation of SAC. Activity measurements show remarkable synergistic effects, forming a volcano-shaped plot over a range of bulk Re mole fractions with a peak at a fraction of 0.4 – 0.5, which corresponds to a 20-fold enhancement of activity over either monometallic catalyst. This intermediate catalyst composition exhibiting optimum activity strongly suggests a bifunctional effect. Through a consecutive reaction pathway with GBL as an intermediate, the main product is THF with 60 – 75% selectivity. Butanol formation is minimized with decreasing Re mole fraction.

**Monday, May 21, 2018
10:30 AM
Swearingen 3C02**