

Catalytic Steam and Dry Reforming of Methane Using Rh Clusters Supported on Sr-substituted Hexaaluminate

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Introduction

This work reports the synthesis, characterization and performance of steam-reforming catalysts based upon dispersed Rh clusters on Sr-substituted hexaaluminate supports. Although reforming typically uses a Ni catalyst at relatively low temperature, the Rh-on-hexaaluminate approach offers some potential advantages. These advantages include resistance to sintering, surface-area loss, and deactivation in high-steam environments [1]. Additionally, although it is certainly more costly, Rh is a much more active catalyst than is Ni.

Materials and Methods

The hexaaluminates are synthesized with an alumoxane process [2] and the Rh is impregnated with metal-nitrate salts. The resulting structures are characterized using X-Ray diffraction (XRD), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). A stagnation-flow reactor is used to measure catalytic activity. In these experiments, the catalyst is applied to a flat surface that is held at a fixed temperature. Reactive gases (methane, steam, and diluent) impinge on the catalytic stagnation surface. Microprobe mass spectrometry is used to measure gas-phase species profiles in the boundary layer normal to the catalyst surface. These experiments are interpreted with chemically reacting flow models that incorporate elementary heterogeneous mechanisms.

Results and Discussion

As confirmed by electron microscopy and XRD, the Sr-substituted hexaaluminate provides a plate-like support structure that resists sintering and occlusion of the Rh. The Rh-on-hexaaluminate structure is found to be very stable, even in harsh accelerated-aging environments.

Rhodium clusters supported on Sr-substituted hexaaluminates are found to perform well for the steam and dry reforming of methane. This paper provides comparisons between predicted and measured gas-phase boundary-layer profiles. A previously developed detailed reaction mechanism is used. Despite the fact that the mechanism was developed for Rh-Al₂O₃ catalyst-support system, it performs remarkably well for the current hexaaluminate-supported system.

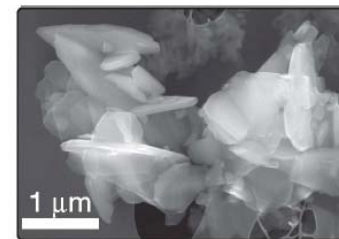


Figure 1. SEM Image of a Sr-substituted hexaaluminate, showing distinct plate-like structure

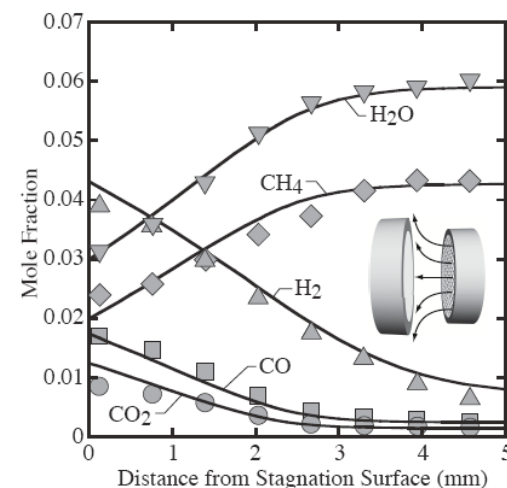


Figure 2. Boundary-layer mole-fraction profiles for a steam-to-carbon ratio of 1.4 and a surface temperature of 740 °C (baseline case)

Significance

The concerted experimental-modeling effort provides new fundamental data and insight concerning the structure and performance of hexaaluminate-supported catalysts. These results can be used in the design and development of catalytic reforming technologies in other geometric configurations.

References

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2. Karevia, A., Harlan, C.J., MacQueen, D.B., Cook, R.L., and Barron, A.R. *Chem. Mater.* 8, 2331 (1996)