

## Stability of hexaaluminate-based materials for catalytic combustion prepared by the microemulsion technique

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### Introduction

One of the biggest challenges with catalytic combustion for gas-turbine applications is to develop materials that have high enough activity and stability for operation in the combustor. No material has shown to be able to operate in the whole range of temperatures in the combustor and the catalyst is therefore divided into at least two segments; the ignition and the high-temperature segment. The high-temperature segment must ignite at the outlet temperature of the ignition segment, which must be limited in order to keep deactivation to a minimum. Hence, the high-temperature segment must be very active and retain the activity during a normal service cycle (>8000 hours). The long operation time implies the use of highly stable materials. One of the most promising materials in terms of sintering resistance is La-hexaaluminate (LHA) [1]. By substituting a few aluminum ions with e.g. manganese, the catalytic activity increases without influencing the sintering resistance [2]. One of the most active hexaaluminates is Mn-substituted La-hexaaluminate (LMHA) [2]. The stability of LMHA is impressive, but it is doubtful whether the activity is high enough to ignite the fuel mixture after the ignition segment. We have therefore searched for alternatives to enhance the activity while keeping the stability of the hexaaluminate. One way is to use the hexaaluminate as a support for more active components. Previous work [3] has shown that hexaaluminate prepared by a conventional water-based method was not suitable as support for LaMnO<sub>3</sub>. By preparing the hexaaluminate using the microemulsion-technique a substantially higher activity was obtained. In the present work we have expanded the previous study to include ceria as active component. The objective of the present study was to investigate the activity and long-term stability of these materials compared with LMHA. We have used the microemulsion technique both to prepare the support and add the active components. By applying this technique it is possible to control the morphology of the support and the degree of interaction between support and active component.

### Materials and Methods

Lanthanum hexaaluminate (LHA) was prepared using the microemulsion technique following the carbonate co-precipitation method. The microemulsion system used was similar as used by Palla et al. [4]. The precipitated particles suspended in the microemulsion were either directly recovered or subjected to hydrothermal treatment at 150°C. After recovery of the particles the materials were calcined at 1200°C. The active components CeO<sub>2</sub> and LaMnO<sub>3</sub> were added in the form of metal nitrates either to the calcined support following the incipient wetness method or as a part of the preparation of the support using the microemulsion technique. The activity of the catalysts was determined by a monolith reactor and. Aging of the catalysts was performed in two steps; accelerated aging at 1100°C for 100 hours in air with 15% steam in order to identify the most promising catalysts and in-situ aging in the reactor for up to 400 hours. The catalyst materials were characterized with XRD, BET and TPR.

### Results and Discussion

By loading 20 wt% of LaMnO<sub>3</sub> or CeO<sub>2</sub> on LHA a higher activity compared to LMHA was obtained as seen in Figure 1. LaMnO<sub>3</sub> had a higher activity than CeO<sub>2</sub> in the lower temperatures, but at higher temperatures they were similar. However, LaMnO<sub>3</sub>/LHA was severely deactivated after the accelerated aging at 1100°C. It can probably be related to migration of Mn into the LHA lattice as observed by XRD. Initial results from CeO<sub>2</sub>/LHA on the other hand indicate a remarkable stability. Even after extended calcination at 1200°C a material with a surface area exceeding 15 m<sup>2</sup>/g was obtained. This can be compared with unsupported CeO<sub>2</sub> that after the same treatment has a surface area of less than 1 m<sup>2</sup>/g. Furthermore, TPR measurements indicate that lanthanum from the support interact with CeO<sub>2</sub>. It is previously known that lanthanum can increase both activity and stability of unsupported CeO<sub>2</sub>. The results from the full aging campaign will be presented at the meeting.

### Significance

One of the key issues with the development of catalytic combustion for gas turbines is the development of high-temperature materials. It is absolutely necessary to verify the aging behavior of the catalyst materials under realistic conditions.

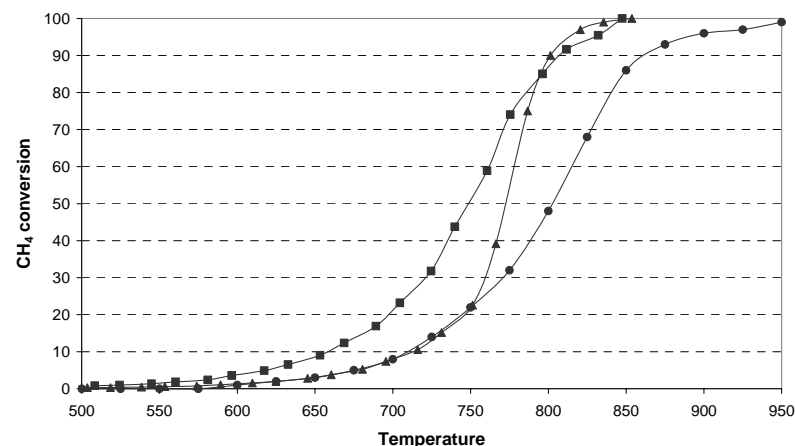


Figure 1. Conversion of CH<sub>4</sub> for 20% LaMnO<sub>3</sub>/LHA (■), 20% CeO<sub>2</sub>/LHA (▲) and LMHA (●). Activity test performed in a monolith reactor, 1.5% CH<sub>4</sub> in air, GHSV = 100,000 h<sup>-1</sup>.

### References

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4. B.J. Palla et al., *Journal of Nanoparticle Research* 1, 215 (1999).