

Hexaaluminates in the catalytic combustion of methane: effect of the deposition over alumina and the addition of palladium

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Introduction

The combustion reaction is frequently used for energy generation. It requires high temperatures and leads to the emission of nitrogen oxides. Catalytic combustion, on the other hand, presents an interesting alternative as it reduces the temperature of combustion as well as the emission of nitrogen oxides, carbon monoxide and UHC which are harmful to the environment [1]. As catalysts are exposed to high temperatures they must have high thermal stability and promote a low ignition temperature [2]. In this work, catalysts based on hexaaluminate $\text{La}_{0.2}\text{Sr}_{0.3}\text{Ba}_{0.5}\text{MnAl}_{11}\text{O}_{19}$ were prepared using the coprecipitation method and deposited on alumina. After the preparation of the hexaaluminate it was supported on γ -alumina and impregnated with palladium acetylacetonate to improve the catalytic activity.

Materials and Methods

Synthesis of $\text{La}_{0.2}\text{Sr}_{0.3}\text{Ba}_{0.5}\text{MnAl}_{11}\text{O}_{19}$ (HX) and $\text{PdO}/\text{La}_{0.2}\text{Sr}_{0.3}\text{Ba}_{0.5}\text{MnAl}_{11}\text{O}_{19}$ (PHX)

The precursor nitrates of La, Sr, Ba, Mn, Al were dissolved in distilled water. Then this solution was added to a solution of ammonium carbonate heated to 60°C and maintained at pH= 8. This mixture was kept under slow agitation for 4 hours. The precipitate was filtered, washed with distilled water, dried at 120°C for 12h and calcined at 1000°C for 6h. The hexaaluminate obtained was mixed in toluene and remained under agitation for 24 h. This was followed by the addition of acetylacetonate of palladium and remained under constant agitation for a further 24 h. After drying at 120°C for 12h, the catalyst was calcined at 650°C for 5h.

Synthesis of $\text{La}_{0.2}\text{Sr}_{0.3}\text{Ba}_{0.5}\text{MnAl}_{11}\text{O}_{19}/\text{Al}_2\text{O}_3$ (HXA) and $\text{PdO}/\text{La}_{0.2}\text{Sr}_{0.3}\text{Ba}_{0.5}\text{MnAl}_{11}\text{O}_{19}/\text{Al}_2\text{O}_3$ (PAHX)

γ - Al_2O_3 was added in a solution of NH_4OH at pH 8 heated to 60°C under intense agitation. Then the solution containing the precursors nitrates of La, Sr, Ba, Mn, Al was added slowly resulting in a pH= 8. The heating and agitation was maintained for 30 min. The precipitate was filtered and washed with distilled water, dried at 120°C for 12h and calcined at 1000°C for 6h. Part of the precursor was impregnated with palladium using the previously mentioned procedure. A catalyst 1% $\text{PdO}/\gamma\text{-Al}_2\text{O}_3$, Al_2O_3 (PA) which was used as reference sample was also prepared by wet impregnation.

Results and Discussion

The XDR of the catalysts revealed the presence of the hexaaluminate phase as well as the presence of the PdO with a tetragonal structure for the catalysts impregnated with the acetylacetonate of palladium. The results of the TG (Figure 1) show that the deposition of the hexaaluminate on the alumina leads to a lower loss of weight to hexaaluminate increasing the thermal stability of these catalysts.

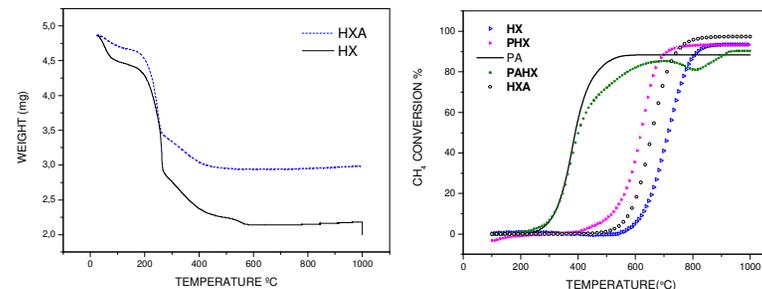


Figure 1- TG of the catalysts HX and HXA

Figure 2- TPRS of the catalysts

According to the TPRS of the hexaaluminate HX (Fig.2) the addition of palladium, PHX, led to a drop in the ignition temperature. The ignition temperature for 10% of conversion of methane of the catalyst HX was 576°C compared to 315°C of the catalyst PHX with palladium. This observation is in agreement with literature [3] that reports the high thermal stability of hexaaluminates but its lower activity compared to palladium oxide catalysts.

The TPRS profiles of the catalysts HX and HXA indicate that HXA was more active in the combustion reaction. Comparing the palladium catalysts PHXA and PHX, it was observed that the activity was more favored over the catalyst PHXA, deposited on $\gamma\text{-Al}_2\text{O}_3$. This can be justified by the presence of alumina which has a higher surface area than the hexaaluminate [2], which would increase the dispersion of active sites favoring the kinetics of the reaction. Among all the catalysts prepared PAHX showed the highest activity with results similar to the ignition temperature of the reference sample 1% $\text{PdO}/\text{Al}_2\text{O}_3$ (PA). PAHX catalysts also presented the $\text{PdO} \rightarrow \text{Pd}$ conversion at 700°C and $\text{Pd} \rightarrow \text{PdO}$ at 800°C.

Significance

In this work the preparation routes used improved catalytic properties of hexaaluminates. The deposition of $\text{La}_{0.2}\text{Sr}_{0.3}\text{Ba}_{0.5}\text{MnAl}_{11}\text{O}_{19}$ over $\gamma\text{-Al}_2\text{O}_3$ improved both thermal stability and the activity on the catalytic combustion. Furthermore, the addition of palladium to the hexaaluminate/ $\gamma\text{-Al}_2\text{O}_3$ decreased the ignition temperature and the activity obtained is similar to the $\text{PdO}/\gamma\text{-Al}_2\text{O}_3$ reference sample.

References

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