

## Study on hexaaluminate $\text{MnLaAl}_{11}\text{O}_{19-\delta}$ catalyst for catalytic combustive reaction of dimethyl ether as a new fuel

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**Abstract:** Study on catalytic combustive reaction of dimethyl ether as a new fuel was presented. Hexaaluminate catalysts were used to reduce ignition temperature so that dimethyl ether completely combusted at low temperature. Hexaaluminate catalysts  $\text{MnLaAl}_{11}\text{O}_{19-\delta}$  were prepared by reverse microemulsion method. Crystalline phase and structure of the catalyst were analysed by means of TG-DTA, XRD and BET. The results show that the hexaaluminate is of magnetoplumbite structure when La is taken as mirror plane cation. Hexaaluminate phase is formed slowly via 1050 °C calcined for 4 h and it can keep hexaaluminate phase and high surface area of  $48 \text{ m}^2 \cdot \text{g}^{-1}$  even calcined at 1200 °C for 2 h. Catalytic activity of  $\text{MnLaAl}_{11}\text{O}_{19-\delta}$  was tested in combustion reaction of dimethyl ether. It shows that hexaaluminate is of high activity with  $T_{10\%}$  at 170 °C and almost 100% conversion at 370 °C.

**Key words:** dimethyl ether; catalytic combustion; La-hexaaluminate; reverse microemulsion

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### 1. Introduction

Being the simplest aliphatic ether, dimethyl ether (DME) has no carbon-carbon bonds, and after methane it has the relative low ratio of carbon to hydrogen. Dimethyl ether has a low boiling point ( $-25.1 \text{ }^\circ\text{C}$ ), a high cetane number (55 – 60), and a high oxygen content ( $\sim 35\%$  by weight). It can be ultra-clean-burned and resourceful, which can be made from natural gas, coal or biomass. It makes DME be a diesel substitute due to low soot emission combustion in diesel engines. Moreover, DME is also expected to be used in power generation and home appliances [1]. As such, its combustion property has been studied recently [2-4]. In order to attain a high energy transforming efficiency and low emissions of air pollutants, a

catalyst with excellent ignition activity and thermal stability is urgently needed [5]. Catalysts supported by noble are well known to be active for high temperature combustion. But their commercial exploitation is restricted due to decomposition, poisoning and high cost [6-7]. Therefore, it is necessary to search a kind of catalysts with relatively high activity, good thermal stability and less expensive.

In recent years, the studies of noble catalyst hexaaluminate applied in methane catalytic combustion reaction have been made a great progress [8-10]. Hexaaluminate materials are high temperature combustion catalysts, due to the high thermal stability associated with their peculiar layered structure, for example,  $\beta\text{-Al}_2\text{O}_3$  structure or magnetoplumbite (MP) structure, intercalated by mirror planes in which the large cations such as Ba, La and Sr are locat-

ed. Introduction of transition metal ions such as  $Mn^{2+}$ ,  $Co^{3+}$ ,  $Fe^{3+}$  and  $Ni^{2+}$  or rare earth material  $Ce^{2+}$ , provides good combustion activity without decrease of sintering resistance [11-12]. With the properties to be able to avoid the sintering and deactivation, and with the properties of thermal stability, high activity, highly heat-resistant and high surface area, hexaaluminate is considered to be one of suitable catalysts for combustion [13]. Because there are similar combustion properties between dimethyl ether and methane in combustion [14-16], we could make investigation about hexaaluminate applied in dimethyl ether catalytic combustion.

In the present work, we have utilized reverse microemulsion method with system consisting of n-heptane, emulsifier OP-10, 1-octanol and water to prepare hexaaluminate  $MnLaAl_{11}O_{19-\delta}$ . The obtained sample was characterized with TG-DTA, X-ray, BET and dimethyl ether combustion reactions.

## 2. Experimental

### 2.1. Catalysts preparation

Mn-modified hexaaluminate was prepared with reverse microemulsion method in system consisting of n-heptane, emulsifier OP-10, 1-octanol and water. Mixed metal nitrates solution at a definite mole ratio, polyethylene glycol 600 as dispersant, were added into reverse microemulsion drop by drop at 80 °C with vigorous stirring. And 4 mol·L<sup>-1</sup> ammonia solution and 2 mol·L<sup>-1</sup> urea solution were mixed with a definite volume ration, and slowly added into reverse microemulsion drop by drop at 90 °C with vigorous stirring until pH 9. The mixture was filtered. The obtained solid was washed by ethanol three times and dried using supercritical drying. The resulting solid was calcined at 1050 and 1200 °C for 2, 4 and 8 h respectively.

### 2.2. Characterization techniques

The weight loss and reaction evolution of the sample were determined with a thermal analyzer NETZS STA409PC. The TGA/DTA samples were heated from 25 to 1400 °C at a heating rate of 20 °C. X-ray diffraction patterns of

the samples were obtained by using a Shimadzu XD-3A with Cu K $\alpha$  radiation. The XRD instrument was operated at 30 kV and 20 mA. And the samples were measured with a 2 $\theta$  scanned from 10° to 90° with a scanning rate of 8 (°)·min<sup>-1</sup>. Surface area of the catalysts was measured with Micromeritics Gemini V.

### 2.3. Catalytic activity measurements

Catalytic activities were tested in a fix-bed reactor at atmospheric pressure. About 0.3 g catalyst particles in the size of 0.3 – 0.5 mm was loaded into a  $\Phi$  8 mm quartz micro-reactor. The measurements were performed at the flow rate of mixed gas (DME and air) adjusted to a GHSV of 6000 h<sup>-1</sup>. The compositions of outlet and inlet gas were determined by using an on-line gas chromatography (GC9790) and a thermal conductivity detector (TCD). The conversion rate of DME was calculated by using carbon balance method.

## 3. Results and discussion

### 3.1. TG-DTA result of sample

Fig.1 shows the TGA and DTA curves of sample  $MnO \cdot La_2O_3 \cdot 6Al_2O_3$  prepared by reverse microemulsion method with supercritical drying. An endothermic peak appears at about 481 °C, corresponding to the weight-loss of 15.9%, which is caused by elimination a little water of hydrate and some oxidations. An exothermic peak appears at 845 °C and there is no weight-loss. This phenomenon is attributed to hexaaluminate crystal phases transition at this temperature. But there is no peak of spinel

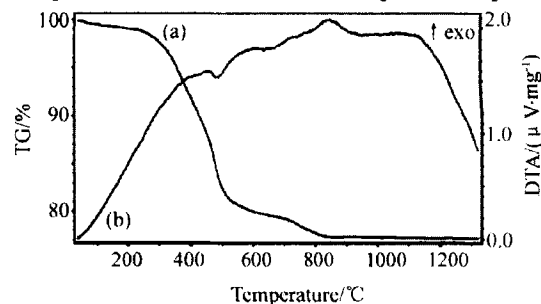


Fig.1. TG-DTA result of sample  $MnO \cdot La_2O_3 \cdot 6Al_2O_3$ : (a) TG curve; (b) DTA curve.

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