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High temperature catalytic combustion of methane and propane over hexaaluminate catalysts: NO_x emission characteristics

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Abstract

Several hexaaluminate-related materials were prepared via hydrolysis of alkoxide and powder mixing method for high temperature combustion of CH_4 and C_3H_8, in order to investigate the effect of the concentration of the fuels, O_2 and H_2O on NO_x emission and combustion characteristics. Among the hexaaluminate catalysts, $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared by the alkoxide method exhibited the highest activity for methane combustion and low NO_x emission capability. NO_x emission at 1500 °C was increased linearly with O_2 concentration, whereas water vapor addition decreased NO_x emission in CH_4 combustion over the $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ catalyst. In the catalytic combustion of C_3H_8 over the $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ catalyst, the amount of NO_x emitted was raised in the temperature range between 1000 and 1500 °C when the C_3H_8 concentration increased from 1 to 2 vol.%. It was found that NO_x emission in this temperature range was reduced effectively by adding water vapor.

Keywords: Hexaaluminate catalyst; Methane combustion; Propane combustion; NO_x

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1. Introduction

Catalytic combustion of hydrocarbons has been extensively investigated as a clean and efficient combustion technology. The number of applications of this technology is increasing in commercial scales; for instance, domestic heating appliance, chemical process heaters, catalytic burners, catalytic cleaner for exhaust gas and volatile organic compounds, and so on. So far, high temperature catalytic combustion of natural gas above 1000 °C has been intensively studied for commercial applications to gas turbine power generation systems, owing to its feasibility of low NO_x emission and high combustion efficiency [1–4]. Besides the applications of such large scales, small scale applications fired with various kinds of fuels other than natural gas will be of great importance in the near future, since co-generation systems, for example microgas turbine systems, will be widely introduced as a distributed power generation system. Even when various kinds of fuels are catalytically combusted in small scales, the features of low NO_x emission and high combustion efficiency must be retained. In combustion of hydrocarbons, which are free of nitrogen-containing compounds, NO_x can be formed mainly due to two distinct mechanisms, thermal NO_x and prompt NO_x [5]. Thermal NO_x formation is significant in the temperature range more than 1500 °C and highly sensitive

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to temperature and oxygen concentration. Prompt NO_x is considered to be released in fuel-rich flame by rapid reaction between radicals and nitrogen via intermediates such as cyanides. The reaction between radicals and N_2 to produce HCN was referred to as the key step for the prompt NO_x formation [6]. In general, the reaction characteristic of prompt NO_x is relatively insensitive to temperature [7].

Hexaaluminate-related compounds have been proposed as a material of high heat resistance [8–11]. The hexaaluminate compounds have the unique layer crystal structure consisting of the alternative stacking of a spinel block and a monatomic layer including large cations such as Ba, Sr, or La. They retain relatively large surface area >20 m^2/g, after heat treatment even at the temperatures more than 1200 °C. Retaining high surface area at high temperatures is of importance for high temperature combustion catalysts, since catalytic materials are sufficiently active at the high temperatures, and therefore the number of active sites determines the combustion characteristics. This heat-resistant feature is closely related to the planar morphology of the hexaaluminate microcrystals, of which crystal growth along [0 0 1] is strongly suppressed. Partial substitution for the Al-site in the hexaaluminate lattice by some transition metals, such as Mn and Fe, leads to enhancement in the catalytic activity for methane combustion without lowering the surface area. One of the successful designs in the hexaaluminate catalyst is $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via alkoxide method [12]. As these active species are well dispersed in the hexaaluminate lattice, severe volatilization or mutual migration of the active species can be avoided. This work aims to investigate NO_x emission characteristics in catalytic combustion of methane or propane over several types of hexaaluminate materials prepared via hydrolysis of metal alkoxide and solid-state reaction. The effect of the concentration of the fuels, oxygen, and water vapor was examined for NO_x emission and combustion characteristics.

2. Experimental

2.1. Catalyst preparation

Hexaaluminate catalysts such as $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$, $Sr_{0.8}La_{0.2}Al_{12}O_{19-\alpha}$, and $BaMnAl_{11}O_{19-\alpha}$ were prepared via hydrolysis of metal alkoxide and powder mixing method. α-Al_2O_3 heat-treated at 1400 °C for 5 h were used as a reference catalyst in combustion tests. A transparent 2-propanol solution of metal alkoxides was obtained by stirring a stoichiometric mixture of strontium or barium (98%, Kishida Chemical) and $Al(OCH(CH_3)_2)_3$ (98%, Kishida Chemical) at 80 °C for 3 h in a nitrogen stream. An aqueous solution of $La(NO_3)_3{\cdot}6H_2O$ (97%, Kishida Chemical) and $Mn(NO_3)_2{\cdot}6H_2O$ (98%, Kishida Chemical), and either $La(NO_3)_3{\cdot}6H_2O$ or $Mn(NO_3)_2{\cdot}6H_2O$, was added to the alcoholic solution of the metal alkoxides, which resulted in precipitate formation by hydrolysis of the metal alkoxides. The precipitate was dried and then decomposed at 500 °C. The resulting precursor was calcined at 1200 °C for 5 h in air. Powder mixing method was applied to prepare $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$. A stoichiometric mixture of commercial powders, $MnCO_3$, $SrCO_3$, La_2O_3, and Al_2O_3 (Kishida Chemical) was mixed and ground in a ball mill for 24 h and then calcined at 1200 °C for 5 h in air.

The surface area of the catalysts was measured by the BET method with N_2 adsorption (Shimadzu, Gemini 2375), and the crystalline phases were identified by X-ray diffraction (Rigaku, RINT1400).

2.2. Catalytic activity test

Catalytic activity of the hexaaluminate catalysts for methane combustion was investigated by using a conventional plug-flow reactor operated at atmospheric pressure. The hexaaluminate catalysts were pelletized, crushed, and sieved to 10–20 mesh. The sieved catalysts (1 ml) were fixed in a quartz tube by packing quartz wool and alumina beads at the end of the catalyst bed. In the catalytic activity test of the hexaaluminate materials, a gaseous mixture of 1 vol.% CH_4, 99 vol.% air was fed to the quartz tube at a space velocity of 48 000 h^{-1}. The gas composition of the feed was varied in methane and propane combustion tests over $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via alkoxide method as follows: CH_4 or C_3H_8, 1–3 vol.%; O_2, 2–20 vol.%; H_2O, 0–6 vol.%; N_2, balance. The effluent gas was analyzed by on-line non-dispersive infrared (NDIR, Shimadzu, GT-7000) and gas chromatography (Shimadzu, GC-8A) to measure the concentration of hydrocarbons, CO and CO_2.

NO_x emission characteristics in catalytic combustion over the hexaaluminate catalysts were evaluated in the temperature range of 900–1600 °C. A conventional flow-type reactor made of an alumina tube (Nikkato, SSA-A) was used for NO_x emission tests by packing the sieved catalysts of 6 ml fixed with perforated alumina plates at the both ends of the catalyst bed. The inlet gas with the same composition as used in the combustion tests was fed to the reactor at a space velocity of 6000 h^{-1}. NO_x concentration at the reactor outlet was measured by a chemi-luminescense method (Shimadzu, NOA-305A).

3. Results and discussion

3.1. Methane combustion and NO_x emission characteristics over hexaaluminate materials

The crystalline phases of hexaaluminate were identified by the XRD measurements for $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via alkoxide hydrolysis and solid-state reaction, $Sr_{0.8}La_{0.2}Al_{12}O_{19-\alpha}$, and $BaMnAl_{11}O_{19-\alpha}$: hereafter abbreviated to SLMA(a), SLMA(b), SLA, and BMA, respectively. The BET surface area of the hexaaluminate materials is summarized in Table 1, along with that of α-Al_2O_3 calcined at 1400 °C for 5 h. The samples prepared via the alkoxide method, SLMA(a), SLA, and BMA, displayed a high BET surface area larger than ca. 15 m^2/g, whereas SLMA(b) showed a low surface area of 0.6 m^2/g. By the solid-state reaction, an intermediate phase such as $BaAl_2O_4$ was formed at first, and then it transformed into the hexaaluminate phase [13]. Thus, it is highly likely that growth of grain particles proceeded during transformation of the intermediate to the hexaaluminate phase, giving rise to the decrease in the surface area of SLMA(b). On the other hand, by the alkoxide method, since fine grain particles were obtained with almost the same composition as in the stoichiometric liquid mixture, the hexaaluminate phase was attained without forming such an intermediate phase. Once the hexaaluminate phase was formed, sintering of the facets was prevented due to the low growth rate of the hexaaluminate materials at elevated temperatures [12]. Consequently, a large surface area was maintained for the hexaaluminate materials prepared via the alkoxide method.

Table 1
BET surface area of hexaaluminate catalysts and α-Al_2O_3

Material	BET surface area ($m^2 g^{-1}$)
$Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via alkoxide hydrolysis (SLMA(a))	17.5
$Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via solid-state reaction (SLMA(b))	0.6
$BaMnAl_{11}O_{19-\alpha}$ (BMA)	14.9
$Sr_{0.8}La_{0.2}Al_{12}O_{19-\alpha}$ (SLA)	25.8
α-Al_2O_3	3.1

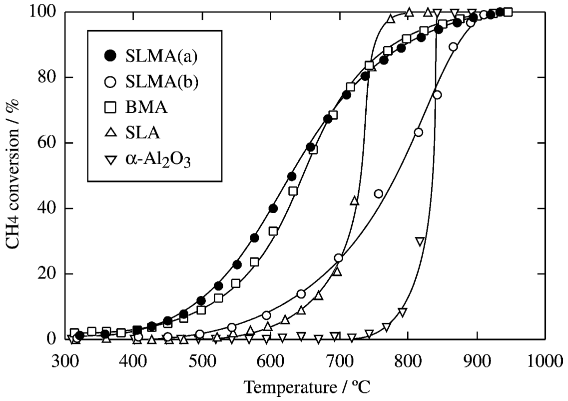

Fig. 1. Catalytic combustion of methane over hexaaluminate materials and α-Al_2O_3: ●, $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via alkoxide hydrolysis (SLMA(a)); ○, $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via solid-state reaction (SLMA(b)); □, $BaMnAl_{11}O_{19-\alpha}$ (BMA); △, $Sr_{0.8}La_{0.2}Al_{12}O_{19-\alpha}$ (SLA); ▽, α-Al_2O_3. Reaction conditions: CH_4, 1 vol.%; air 99 vol.%; space velocity, 48 000 h^{-1}.

Fig. 1 shows methane combustion profile over the hexaaluminate materials and α-Al_2O_3. SLMA(a) and BMA appeared to be active for methane combustion: CH_4 combustion over SLMA(a) and BMA started at around 400 °C, and complete combustion of CH_4 was attained at 900 °C. SLMA(a) showed higher activity than BMA in the temperature range from 500 to 700 °C. Compared to these hexaaluminate catalysts prepared via alkoxide method, SLMA(b) exhibited low methane conversion, especially in low temperature range. It is probable that the small surface area of SLMA(b), as summarized in Table 1, resulted in the low activity for CH_4 combustion. On the other hand, a steep rise in CH_4 conversion up to 100% was observed at 700 and 800 °C for SLA and α-Al_2O_3, respectively. This profile is typical for the homogeneous

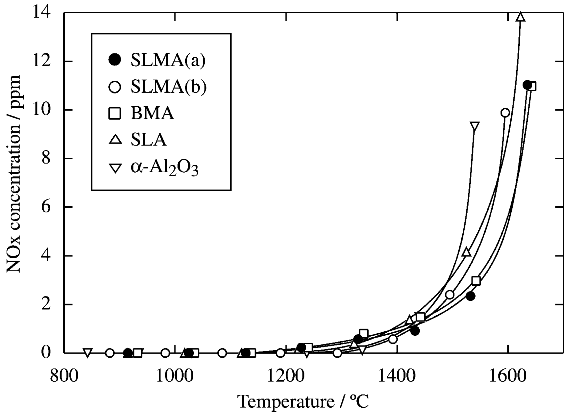

Fig. 2. NO_x emission characteristics in catalytic combustion over hexaaluminate materials and α-Al_2O_3: ●, $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via alkoxide hydrolysis (SLMA(a)); ○, $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via solid-state reaction (SLMA(b)); □, $BaMnAl_{11}O_{19-\alpha}$ (BMA); △, $Sr_{0.8}La_{0.2}Al_{12}O_{19-\alpha}$ (SLA); ▽, α-Al_2O_3. Reaction conditions: CH_4, 1 vol.%; air 99 vol.%; space velocity, 8000 h^{-1}.

combustion of methane in the gas phase. Since SLA and α-Al_2O_3 are lacking in Mn as redox species active for CH_4 combustion, alkyl radicals could be formed on the surface of the materials and transferred to the gas phase, leading to the onset of homogeneous combustion in the gas phase.

NO_x emission characteristics in CH_4 combustion over the hexaaluminate materials and α-Al_2O_3 were measured at high temperatures above 900 °C (Fig. 2). Difference in NO_x emission was distinct at 1500 °C and higher temperatures: NO_x was emitted more over the inactive materials, SLA and α-Al_2O_3, whereas the amount of NO_x formed over SLMA(a) and BMA was smaller. In the catalytic combustion of CH_4 over the inactive materials, it is inferred that alkyl radicals are possibly generated and then released to the gas phase, as discussed above. The alkyl radicals contributed to NO_x formation at lower temperatures compared to SLMA(a) and BMA. On the other hand, because the CH_4 oxidation rate on the surface can be high due to their high activity and thus the CH_4 concentration is low at the surface of SLMA(a) and BMA, less alkyl radicals can be formed at the catalyst surface, leading to the small amount of NO_x emission over SLMA(a) and BMA. As shown above, in addition to the low NO_x emission capability, SLMA(a) exhibited excellent activity for CH_4 combustion and retained high surface area. Therefore, in the following sections, we will focus on the combustion and NO_x emission characteristics over SLMA(a).

3.2. Effect of CH_4, O_2, H_2O concentration on combustion and NO_x emission characteristics

Effect of CH_4, O_2, and H_2O concentration in the feed gas was investigated for catalytic combustion of CH_4 over SLMA(a). Fig. 3(a) shows the CH_4 combustion profiles when the CH_4 concentration was varied between 1 and 3 vol.%. It is clear that CH_4 conversion was independent of the inlet CH_4 concentration. The first order of reaction order with respect to CH_4 concentration was estimated from CH_4 conversion less than 10%. CH_4 conversions are plotted in Fig. 3(b) for O_2 concentration of 2, 5, 10, and 20 vol.%. In the low CH_4 conversion region less than ca. 10%, O_2 concentration appeared to have no influence on the combustion characteristics, while CH_4 conversion was decreased in keeping with O_2 concentration in the high CH_4 conversion region. The decrease was significant especially for the O_2 concentration of 2 vol.%. In the high CH_4 conversion region, mass transfer from the gas phase can be the rate-determining step. Hence, the reaction rate can be reduced for the low oxygen concentrations, leading to the decrease in CH_4 conversion in the high CH_4 conversion region. The reaction order estimated from CH_4 conversion less than 10% was almost zero with respect to O_2 concentration. Fig. 3(c) displays CH_4 conversion profiles for the variation in H_2O concentration of 0, 3, and 6 vol.%. With the addition of H_2O vapor in the feed stream, the onset temperature of CH_4 conversion was shifted toward higher temperature. Thus, the catalytic activity of SLMA(a) was degraded by water vapor, as reported for CH_4 combustion over BMA by Groppi et al. [14]. The influence of water vapor was lessened as the temperature was raised, and the temperature of complete combustion was at around 900 °C independent of the water vapor concentration. CH_4 conversion profiles for the H_2O concentrations of 3 and 6 vol.% were identical, indicating that catalytic activity of SLMA(a) for CH_4 combustion was deteriorated by water vapor to the same extent irrespective of H_2O concentration.

NO_x emission at 1500 °C in CH_4 combustion over SLMA(a) is summarized in Fig. 4 for several CH_4, O_2, and H_2O concentrations. NO_x emission increased

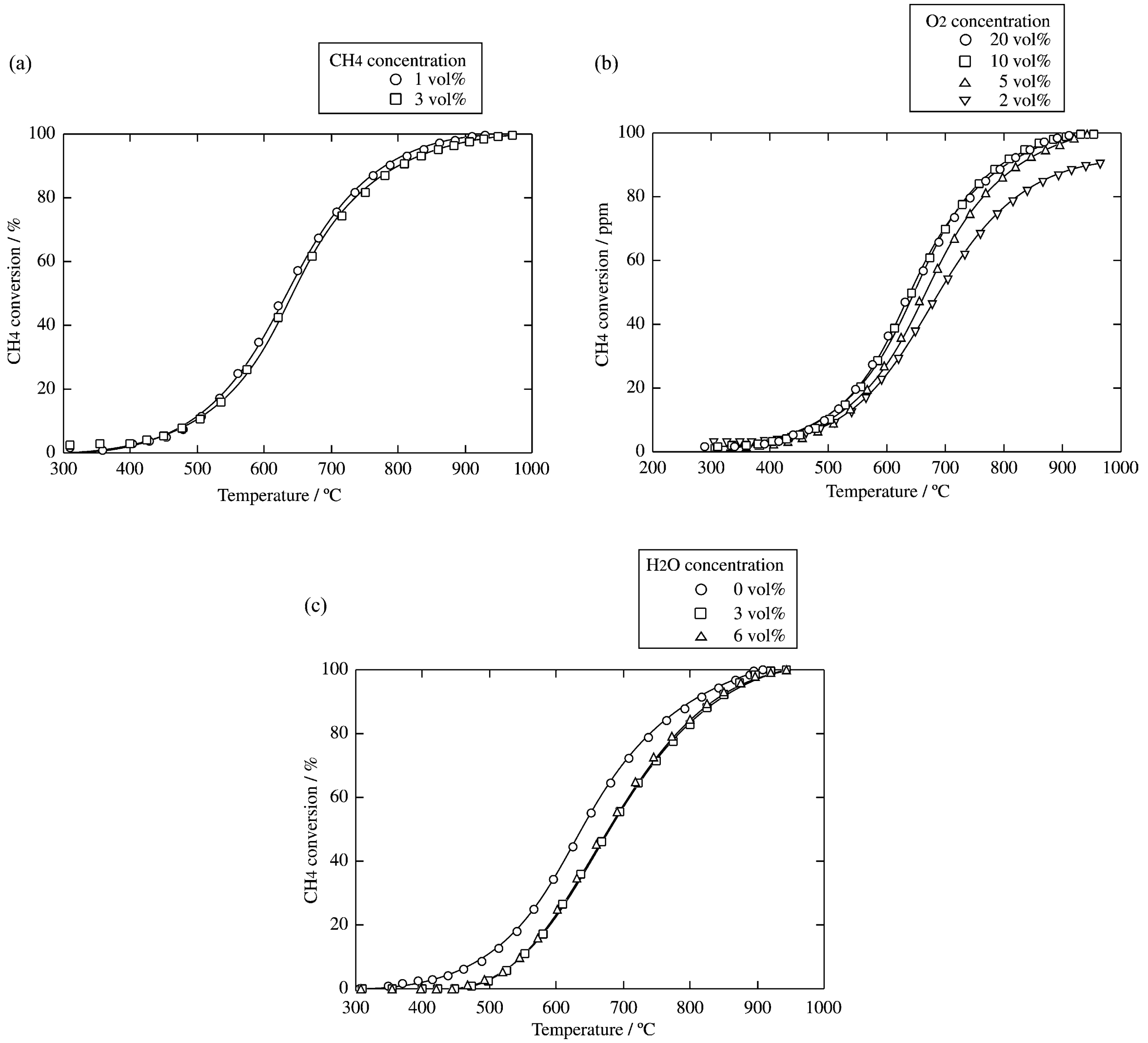

Fig. 3. Effect of (a) CH_4, (b) O_2, and (c) H_2O concentration on CH_4 combustion characteristics over SLMA(a). Reaction conditions: (a) CH_4, 1 or 3 vol.%; air balance; space velocity, 48 000 h^{-1}; (b) CH_4, 1 vol.%; O_2, 2, 5, 10, or 20 vol.%; N_2 balance; space velocity, 48 000 h^{-1}; (c) CH_4, 1 vol.%; H_2O, 0, 3, or 6 vol.%; air balance; space velocity, 48 000 h^{-1}.

almost linearly with oxygen concentration from 2 up to 20 vol.%, while CH_4 had little influence on NO_x emission at 1500 °C in the concentration range of 1–3 vol.%. In general, NO_x is emitted at 1500 °C by thermal NO_x mechanism and thus is governed by the reaction related to oxygen molecular and nitrogen [5]. Therefore, the NO_x emission exhibited direct dependence on oxygen concentration and linearly increased with O_2 concentration, as shown in Fig. 4. When water vapor of 3 or 6 vol.% was added to the CH_4 feed stream, NO_x emission decreased, in reference to the combustion tests without steam. This could be attributed to compensation of intermediates for NO_x formation due to the reaction of them with H_2O [15].

3.3. Propane combustion and NO_x emission characteristics over SLMA(a)

Propane combustion tests over SLMA(a) were carried out to study the effect of C_3H_8, O_2, and steam

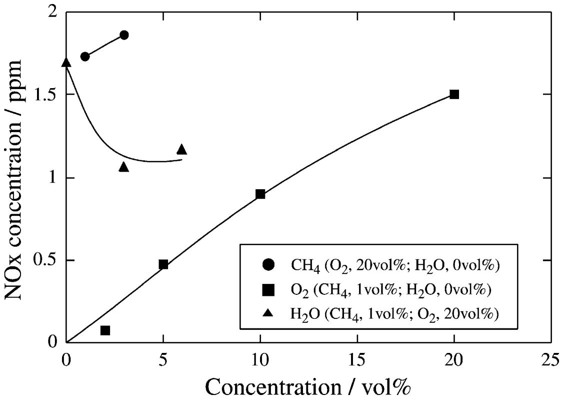

Fig. 4. Dependence of NO_x emission at 1500 °C on the concentration of CH_4, O_2, and H_2O in catalytic combustion of CH_4 over $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via alkoxide hydrolysis (SLMA(a)). Reaction conditions: CH_4, 1 or 2 vol.%; O_2, 0–20 vol.%; H_2O, 0–6 vol.%; N_2 balance; space velocity, 8000 h^{-1}.

concentration on NO_x emission and combustion characteristics. C_3H_8 combustion and NO_x emission characteristics are shown in Fig. 5(a) and (b), respectively. Fig. 5(a) shows that, in comparison with the CH_4 combustion results, C_3H_8 combustion started at lower temperature and C_3H_8 conversion increased more steeply in the low conversion region, and then rose more gradually in the high conversion region. NO_x emission results in Fig. 5(b) exhibited nearly the same trend toward the temperature increase: at the temperatures higher than 1500 °C, a steep rise in NO_x emission was observed, probably due to thermal NO_x formation.

Effect of C_3H_8 and O_2 concentration on combustion characteristics is shown in Fig. 6. The difference in the C_3H_8 conversion was apparent as shown in Fig. 6(a) when the C_3H_8 conversion increased, whereas almost identical conversion profile was attained for C_3H_8 conversion less than ca. 20%. Nearly first reaction order with respect to C_3H_8 was estimated from the conversion less than 10%. The influence of O_2 concentration was considerably different from those observed in the case of CH_4 combustion (Fig. 6(b)). The onset temperature of catalytic combustion of C_3H_8 shifted toward higher temperature as the O_2 concentration decreased, while the difference in the profiles was less significant at the high C_3H_8 conversions, compared to the CH_4 combustion. It is worthy of note that C_3H_8 conversion differed from each other in the low C_3H_8 conversion region, which was not observed for the CH_4 combustion. The reaction order of the C_3H_8 combustion with respect to O_2 concentration was found to be 0.8 ± 0.1.

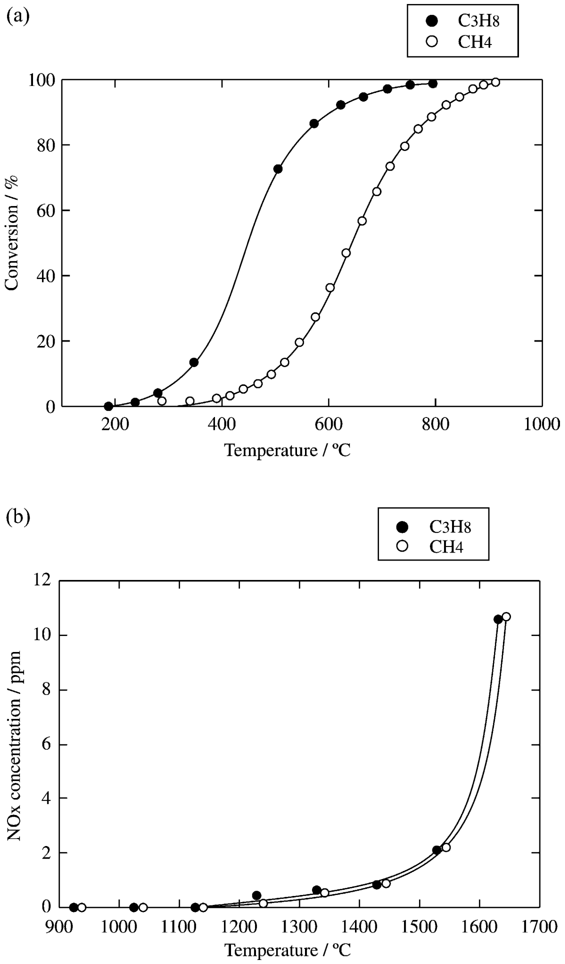

Fig. 5. Catalytic combustion of propane and methane over SLMA(a): (a) combustion characteristics, and (b) NO_x emission characteristics. Reaction conditions: C_3H_8 or CH_4, 1 vol.%; O_2, 20 vol.%; N_2 balance; space velocity, (a) 48 000 h^{-1}, (b) 8000 h^{-1}.

Fig. 7(a) and (b) shows NO_x emissions as the C_3H_8 and O_2 concentrations were varied from 1 to 2 vol.% and from 6 to 20 vol.%, respectively. When the propane concentration increased from 1 to 2 vol.%, NO_x emission characteristics displayed a remarkable change in the temperature range of 1000–1500 °C, as shown in Fig. 7(a). In the case that 1 vol.% propane was combusted over SLMA(a), NO_x emission level remained at the same level below 1 ppm, as the combustion of 1 vol.% CH_4. When 2 vol.% propane was

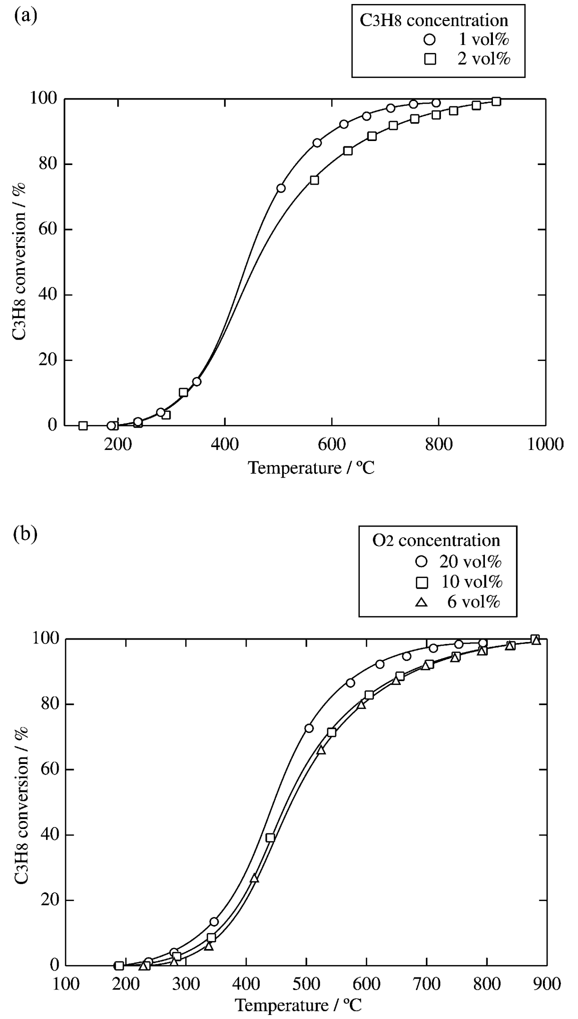

Fig. 6. Effect of (a) C_3H_8 and (b) O_2 concentration on catalytic combustion of propane over SLMA(a). Reaction conditions: (a) C_3H_8, 1 or 2 vol.%; O_2, 20 vol.%; N_2 balance; space velocity, 48 000 h^{-1}; (b) C_3H_8, 1 vol.%; O_2, 6, 10, or 20 vol.%; N_2 balance; space velocity, 48 000 h^{-1}.

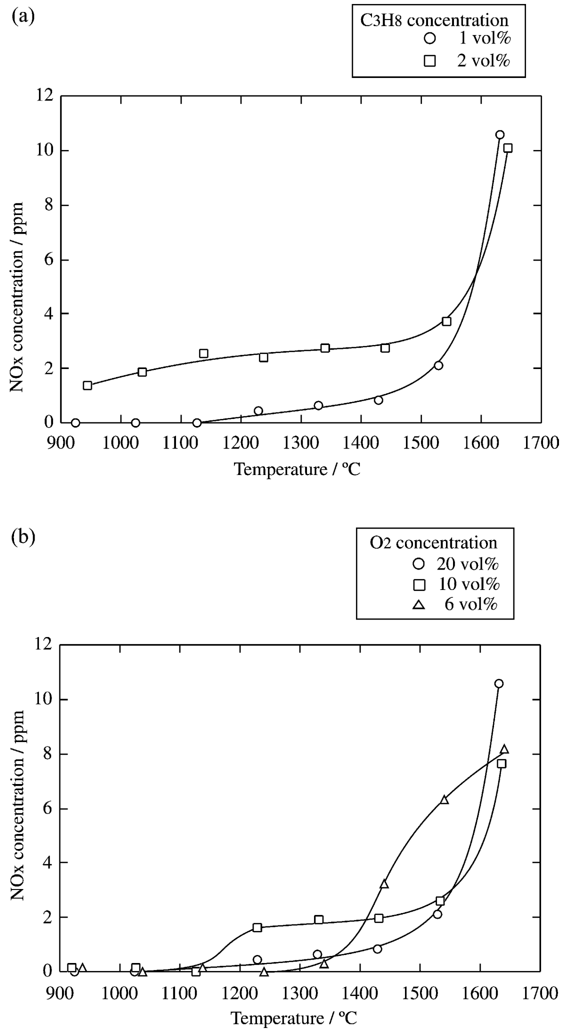

Fig. 7. NO_x emission profiles in catalytic combustion of C_3H_8 over SLMA(a). Reaction conditions: (a) C_3H_8, 1 or 2 vol.%; O_2, 20 vol.%; N_2 balance; space velocity, 8000 h^{-1}; (b) C_3H_8, 1 vol.%; O_2, 6, 10, or 20 vol.%; N_2 balance; space velocity, 8000 h^{-1}.

fed, NO_x emission increased up to more than 2 ppm in the temperature range of 1000–1500 °C. Since the NO_x level emitted in 2 vol.% C_3H_8 combustion was nearly constant and insensitive to temperature in the range below 1500 °C, the increment in the NO_x emission can be attributed to prompt NO_x mechanism where hydrocarbon radicals react with nitrogen. It is inferred that, when the fuel concentration increases, hydrocarbon radicals may be formed more easily in catalytic combustion of C_3H_8, giving rise to the increase in NO_x emission at temperatures lower than 1500 °C. Concerning the influence of O_2 concentration on NO_x emission characteristics, NO_x emission was changed in a complicated manner for the variation in O_2 concentration, as depicted in Fig. 7(b). Compared to the NO_x emission characteristics for the O_2 concentration of 20 vol.%, a rise in NO_x emission was observable in the temperature range of 1200–1500 °C

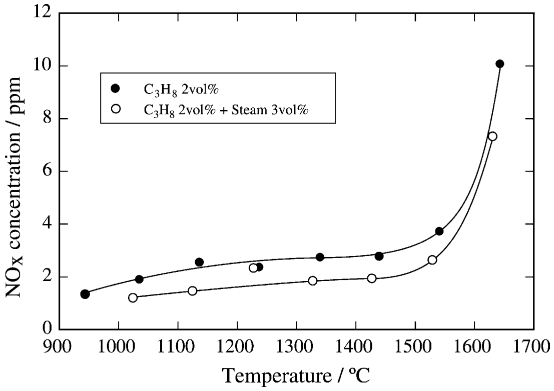

Fig. 8. Effect of water vapor on NO_x emission in C_3H_8 combustion over SLMA(a). Reaction conditions: C_3H_8, 2 vol.%; O_2, 20 vol.%; H_2O, 3 vol.%; N_2 balance; space velocity, 8000 h^{-1}.

in the case of 10 vol.% O_2. On the other hand, when O_2 concentration was 6 vol.%, NO_x emission was higher than the cases with 10 and 20 vol.% O_2, at the temperatures more than 1400 °C.

Fig. 8 illustrates the effect of water vapor on NO_x emission in C_3H_8 combustion over SLMA(a). As shown above, NO_x emission was raised up to more than 2 ppm at temperatures of 1000–1500 °C in the combustion of 2 vol.% C_3H_8. When water vapor of 3 vol.% was introduced to the feed stream containing 2 vol.% C_3H_8, NO_x emission was lowered by 1 ppm in the temperature range of 1000–1500 °C. The effect of water vapor in reducing NO_x emission is more evident in C_3H_8 combustion than in CH_4 combustion. Considering the temperature range and the constancy of NO_x emission in catalytic combustion of C_3H_8, it is likely that the increase in NO_x emission level between 1000 and 1500 °C is related to prompt NO_x formation, and that NO_x formation in the temperature range can be suppressed by steam addition, possibly due to consumption of the intermediates for the NO_x formation by the reaction with H_2O.

4. Conclusions

Several types of hexaaluminate combustion catalysts were prepared via hydrolysis of alkoxide and solid-state reaction and investigated for high temperature methane combustion tests. The hexaaluminate materials prepared via the alkoxide method maintained a relatively high surface area more than 10 m^2/g after heat treatment at 1200 °C, whereas samples via the solid-state reaction exhibited a low surface area. A relationship was found between the catalytic activity for CH_4 combustion and NO_x emission characteristics. When the hexaaluminate catalysts active for CH_4 combustion were used, less NO_x emission was observed at temperatures above 1500 °C, compared to the inactive materials.

Effect of CH_4, O_2, and H_2O concentration on NO_x emission and combustion characteristics was investigated over $Sr_{0.8}La_{0.2}MnAl_{11}O_{19-\alpha}$ prepared via alkoxide hydrolysis (SLMA(a)). No difference appeared in the CH_4 conversion and NO_x emission profiles when CH_4 concentration increased, while the catalytic activity was deteriorated by the increase in H_2O concentration. Nevertheless, suppression effect of water vapor addition on NO_x emission was observed at 1500 °C and higher temperatures.

Propane combustion tests were carried out over SLMA(a). C_3H_8 combustion was initiated at lower temperature than CH_4 combustion. The difference in NO_x emission characteristics between C_3H_8 and CH_4 combustion was negligible when the fuel concentration was as low as 1 vol.%. When the concentration of C_3H_8 was raised to 2 vol.%, an increase in NO_x emission was observed in the temperature range from 1000 to 1500 °C. It was found that the NO_x emission in this temperature range was reduced by adding water vapor, indicating that the rise in NO_x emission in 2 vol.% C_3H_8 combustion could be ascribed to prompt NO_x formation and be suppressed effectively by water vapor addition.

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