Remove cooking fume using catalytic combustion over Pt/La-Al$_2$O$_3$

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Abstract

Five monolithic catalysts with low noble metal content were prepared by immerge method (Pt/γ-Al$_2$O$_3$, Pt/La-Al$_2$O$_3$, Pt/YSZ-Al$_2$O$_3$, Pt+Pd/La-Al$_2$O$_3$ and Pd/La-Al$_2$O$_3$) and their activity measurements were carried out in a conventional fixed-bed flow reactor. The results show that La-Al$_2$O$_3$ can promote activity of the prepared catalysts and can decrease the complete conversion temperature of cooking fume. The Pt/La-Al$_2$O$_3$ catalyst has the highest activity and can be applied in wide range of gas hourly space velocity (GHSV). Some characterizations (XRD, TPR) were carried out with the objective to explain differences in catalytic behaviors. The prepared catalyst showed a great potential for application.

Key words: cooking fume; monolithic catalyst; remove; catalytic combustion

The cooking fume pollution becomes more and more serious with the fast development of the restaurant business. Cooking fume is one of the three pollution sources; it includes many components. At present 99 kinds of compounds have been detected from cooking fume. The prominent components are C$_{16}$ and C$_{18}$ fatty acids (both *n*-alkanoic acids and *n*-alkenoic acids). Cooking fume also includes small amount of polycyclic aromatic hydrocarbons (PAHs). Animal studies have found that PAHs can cause cancer in epithelium of rat cervices (Chiang *et al.*, 1997). Cooking fume has been suggested to increase the risk of lung cancer in oriental women by exposing them to mutagenic substances (Ko *et al.*, 1997). The research results of toxicity have shown that the pollutants in the cooking fume can do a great harm to environment and human health (Benfenati and Pierucci, 1998; Zhou, 1997; Feng and Chen, 2004). It is urgent to control the cooking fume pollution.

Conventional methods for treating cooking fume, such as various types of filters, electrostatic cleaning, mechanical pattern and absorption, have inherent limitations and none is definitely cost-effective (Kornberger and Martin, 2002) The cooking fume conversion efficiency of all these methods can only reach 60%–80%, and going with presents secondary pollution. Therefore, there is currently a great deal of interest in developing the processes that can eliminate cooking fume effectively.

Among the technologies developed for the treatment of cooking fume, catalytic combustion process is considered to be a promising technology. Catalytic combustion has been made to efficiently converter the components of cooking fume to carbon dioxide and water efficiently, at the same time releasing some heat energy (Nishino, 1991; Hobe and Jeffrey, 1996; Davies *et al.*, 1979)

In this study, a series of catalysts with high activity at low-temperatures have been prepared for removing cooking fume. First of all, commercial γ-Al$_2$O$_3$ was used as the catalyst support. To improve the thermal stability of γ-Al$_2$O$_3$, the γ-Al$_2$O$_3$ was impregnated with an aqueous solution of La (NO$_3$)$_3$, and then dried at 120°C for 2 h, calcined at 600°C for 2 h. Thus stabilized γ-Al$_2$O$_3$ contained 3 wt% La$_2$O$_3$(La-Al$_2$O$_3$). The YSZ-Al$_2$O$_3$ (Y$_2$O$_3$-ZrO$_2$-Al$_2$O$_3$) was prepared using a coprecipitation method. Pt was deposited on γ-Al$_2$O$_3$, La-Al$_2$O$_3$ and YSZ-Al$_2$O$_3$ powders by incipient wetness method, respectively. The resulting powders were dried at 110°C for 2 h and calcined at 500°C for 2 h, thus forming Pt/γ-Al$_2$O$_3$, Pt/La-Al$_2$O$_3$, Pt/YSZ-Al$_2$O$_3$ with Pt loading of 0.4 wt%. With the same method mentioned above, Pt+Pd/La-Al$_2$O$_3$ (Pt:Pd = 1:1 (wt)) and Pd/La-Al$_2$O$_3$ were prepared. Desired amount of water was added into the obtained powders to make slurry by ball milling. The resulting slurry was wash-coated onto a monolithic substrate (cordierite honeycomb) with 63 cells/cm^2. The excessive slurry in the cells was removed by compressed air, and dried at 120°C for 2 h, claimed at 500°C for 2 h. The noble metal content on all catalysts was 0.5 g/L.

X-ray diffraction (XRD) were carried out on a DX-1000 X-ray diffractometer with Cu radiation (λ = 0.15418 nm) and graphite filter. The X-ray tube was operated at 45 kV and 25 mA. Samples were scanned from 2θ equal to 10°

up to 90° and the X-ray diffraction line positions were determined with a step size of 0.03° and a slit of 1°.

A micromeritics H$_2$-TPR apparatus was used for the temperature programmed reduction (H$_2$-TPR) analysis. All the samples (100 mg) were pre-treated in a quartz U-tube in an N$_2$ flow at a rate of 30 ml/min (STP). Next, a gas stream of hydrogen (5% H$_2$/N$_2$) was flowed through the sample at a rate of 30 ml/min (STP). The temperature was increased from 30°C to 800°C at a rate of 10°C/min and the amount of hydrogen consumed was determined on a thermal conductivity detector (TCD) as a function of temperature.

The activity measurements were carried out in a conventional fixed bed flow reactor. The gas hourly space velocity (GHSV) of cooking fume was controlled by adjusting the flow rate of passing air into the heated oil (275°C). According to GB 18483-2001, the cooking oil fume effluent from the small column reactor was collected in the impinger filled with 100 ml CCl$_4$ (regent grade) for 10 min (thrice replicates) continuously. The amount of cooking fume was analyzed using JDS-106A infrared oil meter. The amount of cooking fume was determined by the absorption strength at 2930 cm^{-1} (C–H vibration of CH$_2$–), 2960 cm^{-1} (C–H vibration of CH$_3$–), 3030 cm^{-1} (C–H vibration of aromatic rings). The gaseous products were analyzed with an on-line gas chromatograph (GC-2000II) using a Porapak T column, equipped with thermal conductivity detector (TCD) and a flame ionisation detector (FID).

The X-ray diffraction patterns of the as-received catalysts powder samples are shown in Fig.1 That the peaks of Pt are not detected by XRD in the 2θ region from 10° to 50° indicates that the deposited Pt is well dispersed as in the Al$_2$O$_3$ matrix. The diffractograms corresponding to La-Al$_2$O$_3$ do not show diffraction peaks assigned to La$_2$O$_3$ as segregated phases, it is clear that these mixed oxides are solid solutions. At the same time the diffraction peaks of La-Al$_2$O$_3$ are wide, which shows that the crystal is subcrystal. This result is beneficial to promote the catalytic activity. While for Pt/YSZ-Al$_2$O$_3$, the diffractograms corresponding to YSZ-Al$_2$O$_3$ show diffraction peaks assigned to Y$_2$O$_3$-ZrO$_2$ solid solution as segregated phases.
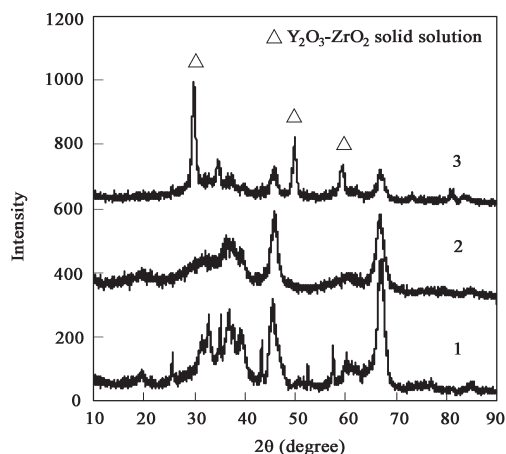
Temperature-programmed reduction has been widely used to characterize reduction behavior of oxide catalysts. PtO$_2$ are well known for its facile reducibility compared with other fluorite-type oxides. The H$_2$-TPR profile of catalysts show a signal pattern, the temperature signal is assigned to the reduction of PtO$_2$. Fig.2 shows the peak temperatures of Pt/La-Al$_2$O$_3$, Pt/YSZ-Al$_2$O$_3$, and Pt/γ-Al$_2$O$_3$ are 239°C, 275°C, 300°C, respectively. The result indicated that the gaseous product was only CO$_2$. Compared with Pt/YSZ-Al$_2$O$_3$ and Pt/γ-Al$_2$O$_3$, peak temperature of Pt/La-Al$_2$O$_3$ shifts toward a lower temperatures, meanwhile the area of the peak of Pt/La-Al$_2$O$_3$ is the largest and peak temperature is the lowest, which indicates that this catalyst may has the best catalytic activity. This fact is in good harmony with the result of catalytic activities in Fig.3.

Fig.3 shows that the results of the catalytic combustion of cooking fume over Pt/La-Al$_2$O$_3$, Pt/γ-Al$_2$O$_3$, Pt/YSZ-Al$_2$O$_3$ catalysts. It is interesting to notice that there is an obvious difference in the catalytic performance of the catalysts for cooking fume. Pt/La-Al$_2$O$_3$ catalyst exhibits higher activity than Pt/γ-Al$_2$O$_3$ and Pt/YSZ-Al$_2$O$_3$. The activity increased in the order: Pt/La-Al$_2$O$_3$ > Pt/γ-Al$_2$O$_3$ > Pt/YSZ-Al$_2$O$_3$. Using Pt/La-Al$_2$O$_3$ catalyst. It is plausible that La-modified alumina can improve the activity of catalyst (Yang *et al*., 2001). So we chose La-Al$_2$O$_3$ in this study as the catalyst support. Different noble metals were deposited on La-Al$_2$O$_3$ and their activities were
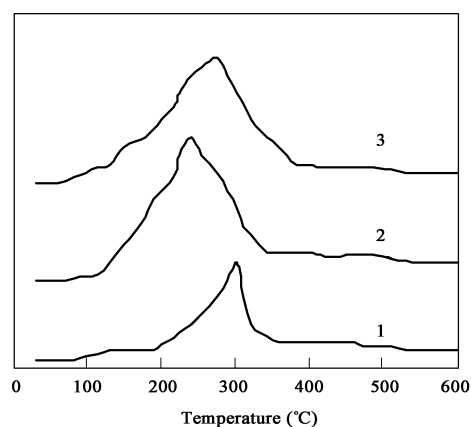
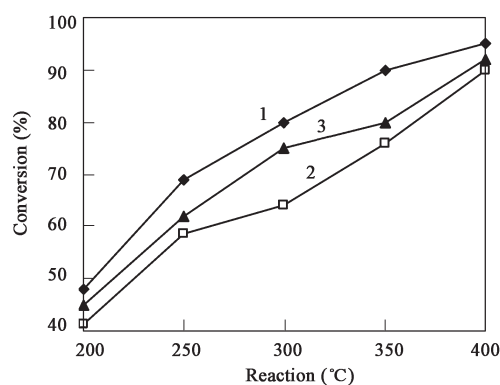Fig. 2 Temperature programmed reduction profiles of catalysts. (1) Pt/γ-Al$_2$O$_3$; (2) Pt/La-Al$_2$O$_3$; (3) Pt/YSZ-Al$_2$O$_3$.

Fig. 1 XRD pattern of different catalysts. (1) Pt/γ-Al$_2$O$_3$; (2) Pt/La-Al$_2$O$_3$; (3) Pt/YSZ-Al$_2$O$_3$.

Fig. 3 Activities of different catalysts. (1) Pt/La-Al$_2$O$_3$; (2) Pt/YSZ-Al$_2$O$_3$; (3) Pt/γ-Al$_2$O$_3$. GHSV = 10000 h^{-1}.

investigated (Table 1).

Table 1 Activity of different noble metals deposited on La-Al$_2$O$_3$

Catalyst	Conversion at different reaction temperature (%)				
	200°C	250°C	300°C	350°C	400°C
Pt/La-Al$_2$O$_3$	48	69	80	90	95
Pt+Pd/La-Al$_2$O$_3$	42	60	74	86	91
Pd/La-Al$_2$O$_3$	30	46	58	68	78

GHSV = 10000 h^{-1}.

It can be seen from Table 1 that the activity order of the three catalysts is Pt/La-Al$_2$O$_3$ > Pt+Pd/Al$_2$O$_3$(1:1) > Pd/La-Al$_2$O$_3$. The Pt/La-Al$_2$O$_3$ shows the highest activity for cooking fume combustion among the tested catalysts. The temperature at 50% cooking fume conversion is found to be 201°C. It is obvious that Pt used as the catalyst active particles can promote activity of catalyst and decrease completed conversion temperature. So, in this study we chose Pt/La-Al$_2$O$_3$ catalyst to investigate the effect of GHSV on the conversion process.
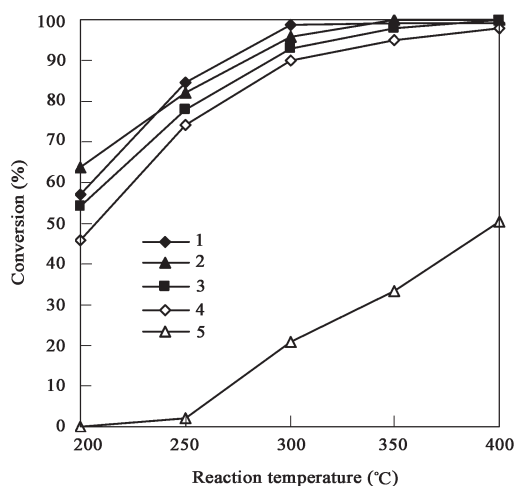

Fig. 4 Effect of GHSV on conversion. GHSV: (1) 10000 h^{-1}; (2) 20000 h^{-1}; (3) 40000 h^{-1}; (4) 60000 h^{-1}; (5) 80000 h^{-1}.

As shown in Fig.4, when increasing GHSV, the activity of catalyst does not change much in the GHSV range of 1×10^4–6×10^4 h^{-1}, while GHSV is 8×10^4 h^{-1}, the activity

of catalyst decreases sharply, the light-off temperature of cooking fume for catalyst increases to 400°C. It is possible that the increase of GHSV shortens the contacting time of the reactant with catalyst. When GHSV is low, the catalysts give more favorable results. So it is obvious that as-prepared catalyst can be applied in wide range of GHSV.

The above results show that the low concentration of noble catalyst of Pt/La-Al$_2$O$_3$ has lower complete conversion temperature and wider operation range of GHSV. It shows a great potential for application. The related experiments on the lifetime of catalyst are underway.

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